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These proceedings contain presentations made at the conference by some of the world's foremost experts in the field of oxidant air quality modeling and presentations made by national experts on their countries' emissions inventories and air quality monitoring activities. Also included are discussions of the presentations, informal presentations, panel discussions, and conference conclusions and recommendations. Among the subjects discussed were the need for and utility of regional oxidant models, six regional models currently under development, and available aerometric and emissions inventory data bases in OECD countries.

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April 12-14, 1983
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Research Triangle Park, North Carolina 27711 (USA)

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The U.S. Environmental Protection Agency (EPA) and the Organization for Economic Cooperation and Development (OECD) are concerned (1) by the fact that the photochemical oxidant pollution problem, due to large-scale formation or long-range atmospheric transport, has international dimensions, and (2) by the lack of ready-to-use methods for formulating optimum control strategies for regional oxidant reduction. In reaction to these concerns, the U.S. EPA and OECD jointly organized the international conference documented in these proceedings.

These proceedings contain presentations made at the conference by some of the world's foremost experts in the field of oxidant air quality modeling and presentations made by national experts on their countries' emissions inventories and air quality monitoring activities. Also included are discussions of the presentations, informal presentations, panel discussions, and conference conclusions and recommendations. Among the subjects discussed were the need for and utility of regional oxidant models, six regional models currently under development, and available aerometric and emissions inventory data bases in OECD countries.

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LIST OF ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

AAQS -- Ambient Air Quality Standard
AGL -- Above ground level
AIRS -- Aerometric Information Retrieval System
AQCR -- Air Quality Control Region
AQMS -- Air Quality Management System
AT -- Air temperature
b-scat -- Light-scattering coefficient
BMC -- b-matrix compiler
BNL -- Brookhaven National Laboratory
CAA -- Clean Air Act
CCN -- Cloud condensation nuclei
CDHS -- Comprehensive Data Handling System
CMA -- Census Metropolitan Area
CMC -- Canadian Meteorological Center
CPU -- Central processing unit
DOC -- Department of Commerce
DOE -- Department of Energy
DPT -- Dew point temperature
ECAO -- Environmental Criteria and Assessment Office

ECE -- Economic Commission for Europe

EIS -- Emissions Inventory System

EIS/AS -- Emissions Inventory Subsystem/Area Sources

EIS/P&R -- Emissions Inventory Subsystem/Permits and Registration

EIS/PS -- Emissions Inventory Subsystem/Point Sources

EKMA -- Empirical Kinetic Modeling Approach

EMEP -- European Monitoring and Evaluation Programme

EMSL -- Environmental Monitoring Systems Laboratory

EPA -- Environmental Protection Agency

EPRI -- Electric Power Research Institute

EPS -- Environmental Protection Service

ERC -- Environmental Research Center

ESRL -- Environmental Sciences Research Laboratory

FRG -- Federal Republic of Germany

GC -- Gas chromatography

GMT -- Greenwich Mean Time

GOES -- Geostationary Operational Environmental Satellite

GWL -- Grosswetterlagen

HATREMS -- Hazardous and Trace Emissions System

HDD -- Heavy-duty, diesel-powered

HDG -- Heavy-duty, gasoline-powered

HDV -- Heavy-duty vehicle

HERL -- Health Effects Research Laboratory

HEW -- Department of Health, Education, and Welfare

HPA -- Heavily polluted area

IERL -- Industrial Environmental Research Laboratory

IVL -- Swedish Water and Air Pollution Research Institute

LDV -- Light-duty vehicle

MAP3S -- Multistate Atmospheric Power Production Pollution Study

MIF -- Model input field

MPO -- Metropolitan Planning Organization

NAAQS -- National Ambient Air Quality Standards

NADB -- National Aerometric Data Branch or National Air Data Branch

NAMS -- National Air Monitoring System

NAPAP -- National Acid Precipitation Assessment Program

NAPS -- National Air Pollution Surveillance

NASA -- National Aeronautics and Space Administration

NATO -- North Atlantic Treaty Organization

NATO/CCMS -- North Atlantic Treaty Organization/Committee on the Challenges of a Modern Society

NAVF -- Norwegian Research Council for Science and the Humanities

NCC -- National Computer Center

NECRMP -- Northeast Corridor Regional Modeling Project

NEDS -- National Emissions Data System

NEROS -- Northeast Regional Oxidant Study

NWS -- National Weather Service

OAQPS -- Office of Air Quality Planning and Standards

OECD -- Organization for Economic Cooperation and Development

ORD -- Office of Research and Development

PAQSM -- Photochemical Air Quality Simulation Model

PEPE -- Persistent elevated pollution episode

PIF -- Processor input file

QSSA -- Quasi-steady-state approximation

RAMC	-- Regional Air Mass Characterization
RAPS	-- Regional Air Pollution Study
RMDHS	-- Regional Model Data Handling System
ROM	-- Regional Oxidant Model
RTI	-- Research Triangle Institute
RTM	-- Regional Transport Model
SAI	-- Systems Applications, Inc.
SAROAD	-- Storage and Retrieval of Aerometric Data
SCC	-- Source classification code
SFT	-- Norwegian Pollution Control Authority
SIC	-- Standard Industrial Classification
SIP	-- State Implementation Plan
SLAMS	-- State and Local Air Monitoring Stations
SURE	-- Sulfate Regional Experiment
TNO	-- The Netherlands Organization for Applied Research
TSP	-- Total suspended particulate
TSR	-- Total solar radiation
UK	-- United Kingdom
USA	-- United States of America
UTM	-- Universal Transverse Mercator
UV	-- Ultraviolet
UVR	-- Ultraviolet radiation
VOC	-- Volatile organic compound
WHO	-- World Health Organization

SYMBOLS

C -- Carbon

HC -- Hydrocarbon

NMHC -- Nonmethane hydrocarbon

NO -- Nitric oxide

NO_x -- Nitrogen oxides

NO_2 -- Nitrogen dioxide

NO_3 -- Nitrate

O_3 -- Ozone

PAN -- Peroxyacetyl nitrate

PBN -- Peroxybenzyl nitrate

SO_x -- Sulfur oxides

SO_2 -- Sulfur dioxide

SO_4 -- Sulfate

TNMHC -- Total nonmethane hydrocarbon

INTRODUCTION

Alfred H. Ellison

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The EPA Office of Research and Development (ORD) is composed of a headquarters located in Washington, DC, and 14 laboratories located throughout the United States. Four of these laboratories are located at the Environmental Research Center (ERC) in the Research Triangle Park.

The major focus of ERC is air pollution research, although other environmental research is conducted. The four laboratories at ERC are the Health Effects Research Laboratory (HERL), the Environmental Monitoring Systems Laboratory (EMSL), the Industrial Engineering Research Laboratory (IERL), and the Environmental Sciences Research Laboratory (ESRL).

Most of HERL's research in health effects involves animal studies. However, HERL also operates a clinical facility in Chapel Hill in which human subjects are placed in a chamber, exposed to pollutants, and monitored for exposure effects.

EMSL is responsible for monitoring activities related primarily to air pollutants. Recently, EMSL has conducted a lot of work in hazardous waste monitoring.

IERL's focus is the development of control systems that are needed for stationary sources of air pollution. As a laboratory, they are concerned with assessing source pollutants and the control systems available for these pollutants. Where there are difficiencies, IERL attempts, at least, to develop prototype systems for controlling air pollution from stationary sources.

ESRL conducts mostly air pollution research and atmospheric sciences research, which brings us to the subject of this conference.

In addition to the four laboratories, several other organizational units are located at ERC. One is the Office of Administration, which provides administrative support to ERC. Another is the National Computer Center (NCC), which includes the two large computers used in EPA's data processing activities. The Office of Air Quality Planning and Standards (OAQPS) is primarily responsible for writing the regulations related to air pollution. Finally, the Environmental Criteria and Assessment Office (ECAO) produces criteria documents. These documents, which are primarily air pollution documents, are produced approximately every 5 yr and review all of the available information on a particular pollutant or toxic chemical.

Alfred Galli

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Good morning. Speaking as a member of the OECD AMP Group the and U.S. EPA-ORD staff, I am delighted that the Group's efforts in the photochemical oxidant control area have culminated in this extremely useful project and that the research staff of the U.S. EPA will have the opportunity to be of assistance. Reflecting ongoing concerns of the U.S. EPA, the past and present U.S. representatives in the AMP Group have repeatedly expressed interest in the development and use of regional-scale models for pollution control strategy development purposes. The long-range transport of polluants is now an established fact and has consequences of concern both within a country, such as the U.S., and to clusters of countries, such as Europe, North America, etc. To understand such problems and to design for each of the involved countries effective and equitable efforts for their control, it is imperative that regional models be used as tools. However, regional models, especially those for oxidants, as you will hear repeatedly in this conference, are extremely difficult to develop and use, and they require substantial commitments on the part of the interested countries.

The United States is certainly an interested country. We are interested because we experience both domestic and international problems associated with long-range pollutant transport. As a result, we have expended and continue to expend substantial resources in efforts to solve these problems, and we would certainly welcome the opportunity to pool our resources with those of other countries for that purpose.

Pierre Lieben, Secretary

Organization for Economic Cooperation and Development
AMPG/Environmental Directorate
Paris, France

This workshop has been designed to comparatively examine existing models for long-range ozone transport with respect to their conceptual validity, complexity, and input data as well as with respect to resource requirements for evaluating and subsequently utilizing one or more of the models in order to eventually devise a strategy for OECD member countries to satisfactorily control the photochemical oxidants pollution problem on an international scale. Such a plan should address the following points:

- Determine the state of the art of existing emissions inventories and develop a plan to complete these as model input data.
- Refine the best available model(s) that can be used with those data bases that appear to be easily obtainable.
- Discuss the merits of selected model(s) with respect to their serving as bases for developing control strategies.
- Recommend a chemistry to be used in the suggested model(s) that best fits the input data likely to be available.
- Examine whether the aerometric data base (including boundary condition data) is sufficient and, if not, formulate a plan to get the missing data.
- Identify dates at which the model(s) will be fully operational.
- Define the modeling domain in consideration of the emissions and aerometric data available, the topography, and the model capability.

It is hoped that during Session VI the above questions will be answered. It is suggested that this outline form the basis for discussions during the meeting, especially in Section IV.

Basil Dimitriades

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To the introductory remarks offered by the preceding speakers, I wish to offer a few comments on the rationale behind the conference agenda, in the hope that this will further help in focusing the conference discussions. These comments are based on the extensive experiences of the U.S. EPA staff with physicochemical modeling in general and with regional O_3 modeling in particular. They are intended to convey the message that in order for the conference to be successful, it is crucial that the conferees go away with a realistic appreciation of the complexity of comprehensive regional O_3 models, and of the penalties and merits associated with model simplification. Crucially important also is the need for the conferees to appreciate the skill and effort requirements of a regional O_3 model evaluation effort, such as the one contemplated by OECD.

The U.S. EPA has been involved in physicochemical modeling of air quality for almost a decade. Model development efforts started in earnest in 1974 when the Regional Air Pollution Study (RAPS) was initiated in St. Louis for the purpose of providing a field data base for modeling urban air quality. The emphasis at that time was on urban-scale modeling, and the efforts eventually resulted in several urban-scale PAQSMs for O_3, some of which were extensively evaluated by the U.S. EPA using the RAPs data base. From these experiences with the urban models, it became apparent that the most useful and valid physicochemical models, that is, those with the highest degree of spatial and temporal resolution and with detailed chemistry, should also have extremely high skill and resource requirements. To illustrate the latter problem, for each of these models now in existence, there are currently only two to three institutions at the most in the United States that have demonstrated their ability to use the model with confidence.

Development of regional PAQSMs was initiated in the late 1970s after the air pollution scientists realized that air pollutants can travel long distances and that photochemical smog problems cannot necessarily be solved or alleviated by controlling local emissions only. These realizations led to the conclusion that for control strategies to be rational, it is imperative that the role and contribution of distant upwind sources be considered, and prompted the U.S. EPA to launch an extensive regional model development program.

Unlike the approaches taken by others, the approach taken by the U.S. EPA modelers to regional-scale modeling was not based on simple expansion of urban-scale models. Such an approach is not apropriate for truly regional models since it is limited to treating only those physical processes that occur or are important during a single solar day and within a few miles from the source. The U.S. EPA regional model currently under development will treat, in addition, nighttime chemistry, very slow reactions, biogenic organics chemistry, air movements associated with the nocturnal jet, cumulus cloud effects, and the processes and effects that have significance mainly in connection with multiday pollutant transport. Thus, relative to other regional modeling approaches, this one approved by the U.S. EPA is conceptually more valid. However, it is also enormously more complex, difficult, and costly to use.

These comments on the differences among the various regional modeling approaches are offered for the purpose of pointing out two implications bearing on the conference conclusions and followup decisions. The first implication is that, for a given regional model application, the most detailed and conceptually valid model is not necessarily the most appropriate and desirable choice. Equally useful may be models that have somewhat lower validity but are much less difficult and costly to use. It is precisely for the purpose of helping the conferees judge the existing models from such standpoints that we included in the agenda presentations on the needs and intended applications of regional O_3 models in the United States and in Europe. I urge the conferees to consider carefully these presentations in their deliberations regarding the relative merits and limitations of existing models with respect to serving the OECD needs.

The second implication is that the conferees should come away with a clear and realistic appreciation of the differences among the various existing models with respect not only to scientific validity but also to practicality. In the hope of facilitating comparisons of models, we have requested that the modelers describe their models following given, detailed guidelines (questionnaires). Adhering to these guidelines will certainly ensure a measure of comparability, but it is questionable whether these standardized model descriptions alone will allow useful judgments to be made upon the practiciality aspect. Such information will have to be extracted from those with extensive experience in the evaluation and application of physicochemical models. It is for this purpose that we have included in Sessions IV and V of the conference panel discussions addressed to, among other things, resources needed for model evaluation. Such discussions should deal not only with money and man-years but also expertise requirements. Only for the purpose of illustrating this point, I offer some very rough estimates made by the U.S. EPA modelers for testing the U.S. EPA regional model with European field data. Such an effort, encompassing processing of European emissions and aerometric data, revising the model to increase its domain (to cover the European OECD-member countries) and to allow for varying terrain roughness (several European countries are predominantly mountainous), and running the model, is estimated to be roughly three expert man-years or more. Since the U.S. EPA staff with working experience in the regional modeling area consists of three persons, it follows that such a model-testing project would require the full-time involvement of the entire U.S. EPA regional modeling capabiliy for one year or more. Although these estimates are very rough, they nevertheless illustrate the point that such a project would require almost prohibitively large commitments in resources It is crucial, therefore, that the conferees appreciate such problems and seek compromises that entail realistic model testing efforts and also adequately serve the needs of OECD.

My last comment is to stress that we at the Environmental Sciences Research Laboratory of the U.S. EPA are truly delighted to have this opportunity to discuss with our international colleagues this extremely important subject. We look forward to the presentations and followup discussions, and we feel committed to assisting OECD in any way we can in this worthwhile undertaking.

We have an enormous respect for our international colleagues and we would be anxious to continue the close contacts that we expect to make during this conference. In this vein of enthusiasm, I would like to submit for consideration by the U.S. EPA and OECD that the U.S. EPA host within the ESRL facility one or two OECD modelers to work with us for a period of one year or so in the regional modeling area. We would certainly gain much from such a close exposure to our guest experts, and OECD would enhance its capability for conducting future modeling projects, such as the one contemplated as a followup to this conference.

My wishes to you all for a productive and enjoyable meeting.

SESSION I

EXISTING REGIONAL MODELS FOR OXIDANTS

April 12, 1983

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NEEDS AND APPLICATIONS OF REGIONAL AIR QUALITY SIMULATION MODELS FOR OXIDANTS IN NORTH AMERICA*

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INTRODUCTION

The purpose of this paper is to discuss the needs and applications of regional air quality simulation models for oxidants in North America. The paper focuses on O_3, the oxidant designated in the National Ambient Air Quality Standard (NAAQS), and on the northeastern United States, the area that is being modeled by the U.S. Environmental Protection Agency (EPA).

Ozone is one of the most serious and widespread air pollution problems in the United States. About one-third of the nation's population lives in 32 urban areas that have been designated as nonattainment regions for O_3. Under the Clean Air Act Amendments of 1977, states have primary responsibility for developing and enforcing programs to control O_3. In accord with this responsibility, the states in 1982 issued plans that describe how they intend to achieve the NAAQS for O_3 by 1987, the statutory deadline. These State Implementation Plans (SIPs) are currently under review by EPA (1983).

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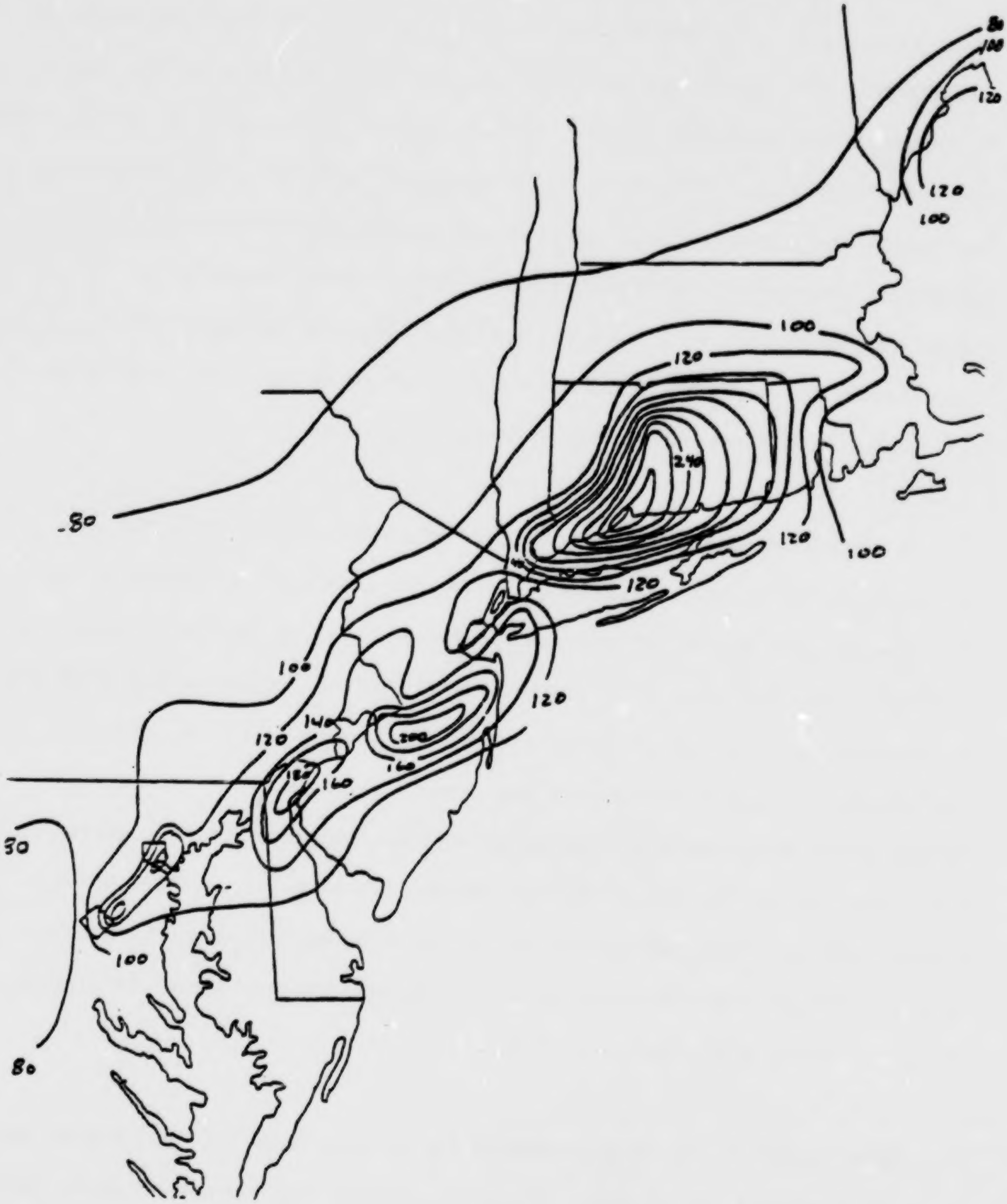
*This paper has been reviewed by the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

The problem of interurban/regional transport of O_3 and/or precursors was first recognized in the Los Angeles Basin during the 1950s when the impact of Los Angeles emissions on the San Bernadino and Riverside areas of the Eastern Basin was established (HEW, 1970). During the past 10 yr, numerous studies have demonstrated the significance of O_3 and precursor transport. The maximum O_3 concentrations associated with urban source regions are recorded tens of kilometers downwind of the source regions (Martinez and Meyer, 1978). Significant transport of O_3 over hundreds of kilometers has also been demonstrated in the Midwestern United States (White et al., 1976, Vukovich, 1977) and in the Northeastern United States (Cleveland et al., 1976).

Several studies present evidence for interstate transport in the Northeastern United States. Figure 1 shows the isopleths of maximum O_3 observed on a day when the winds were from the southwest, i.e., along the metropolitan corridor. The very high O_3 concentrations in Connecticut (about 100 km downwind of New York City) appear to be associated with morning rush hour emissions from the tri-state metropolitan area of New Jersey, New York, and Connecticut. During an EPA study based on Lagrangian aircraft measurements, the Baltimore O_3 plume was tracked for 400 km into New England (Clark et al., 1982). Other studies indicate that substantial quantities of O_3 are periodically transported from the Midwest and Gulf State cities to the Northeastern States (Clarke and Ching, In press; Wolff and Lioy, 1980).

Unfortunately, the legal framework for O_3 control is not well suited to the regional nature of the problem. The responsibility for attainment rests with individual states, and there is no formal mechanism for multistate control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120
200
160
160
180
160
80
100
80

Figure 1. Isopleths of maximum observed O_3 concentrations (ppb) for July 16,1980.

programs. Secondly, regional oxidant models have not been available to the states for the 1982 SIPs. In most cases, the states have used the simple city-specific EKMA Model (EPA, 1981), which treats transport simplistically. In most cases, days with significant O_3 transport were not included in the analyses, the assumption being that control efforts in upwind cities would eliminate this problem in the future. Although this approach appears to be reasonable for areas with moderate O_3 levels, the use of refined regional oxidant models may be desirable for areas that will have difficulty attaining the O_3 NAAQS by 1987.* The experience with O_3 SIPs suggests that a change from the traditional approach to a multistate approach may be required to develop an effective and equitable control program for the Northeast United States.

THE NORTHEAST CORRIDOR REGIONAL MODELING PROJECT

The Northeast Corridor Regional Modeling Project (NECRMP) is a joint program of the Northeastern United States and the EPA (EPA, 1980). Initiated in the late 1970s, NECRMP grew out of the recognition that a serious and widespread O_3 problem exists in the Northeastern United States and that violations are strongly affected by the regional transport of O_3 and its precursor pollutants.

The NECRMP region, shown in Figure 2, encompasses the region from northern Virginia to northern New England and from the East Coast to eastern Ohio and southern Ontario. Five major urban areas--Washington, DC, Baltimore, Philadelphia, New York City, and Boston--are included in the area. The purpose

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*For example, the SIP estimates for NMHC reductions were 60% for New York City and 85% for Los Angeles (EPA, 1983).

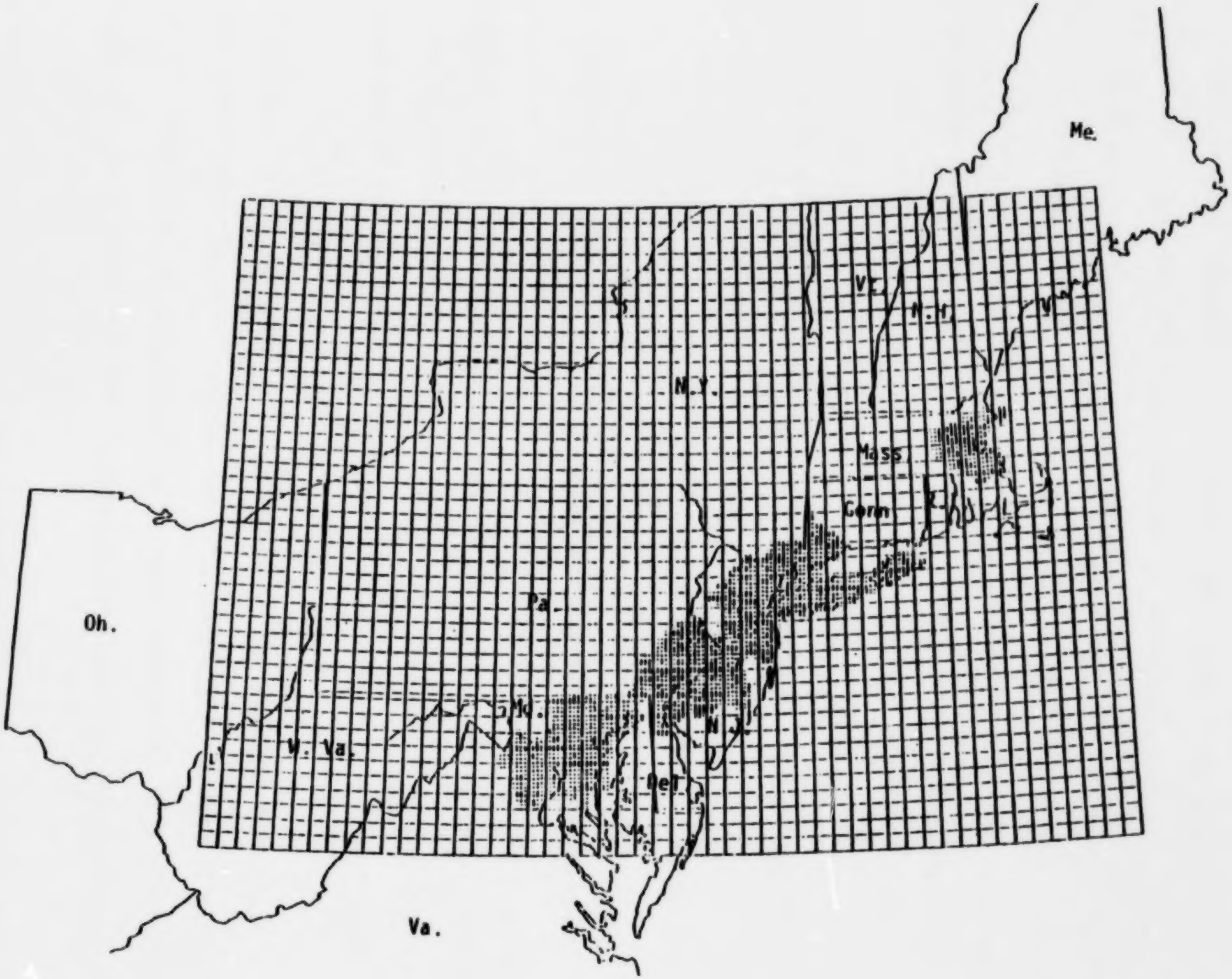
Me.
Vt.
N.H.
Mass
Conn
N.Y.
Pa.
Oh.
W. Va.
Md.
N.J.
Del
Va.

Figure 2. Boundaries of NECRMP domain.

of the project is to use refined photochemical models to develop effective and equitable strategies for O_3 control in the region. As Figure 3 shows, the basic concept of the program is to integrate the use of urban- and regional-scale models.

A key component of NECRMP is EPA's Regional Oxidant Model (ROM) (Lamb, 1982). ROM is a three-dimensional photochemical grid model that simulates the effects of emissions, meteorology, chemistry, and deposition on concentrations of O_3 and other pollutants (CO, NO, NO_2, NMHC, etc.). Its scale enables the model to represent transport between cities and over the full extent of the NECRMP region. Both single-day and multiday simulations are possible. ROM will be used: (1) to ascertain the importance of transport on a scale of 100 km or more, (2) to estimate broad reductions in emissions required to meet the O_3 standard across the Corridor region, and (3) to provide estimates of future-year boundary concentrations that are required as inputs for the urban-scale model applications.

The ROM, however, has too course a grid (18 km x 18 km horizontal cells) to simulate the gradients and the peak concentrations that occur immediately downwind (within 100 km) of urban areas. Thus, the NECRMP concept incorporates the use of an urban-scale photochemical grid model (i.e., the Urban Airshed Model) to develop strategies that will ensure attainment in the major urban areas. Initial plans called for modeling in all five of the major urban areas. However, limited resources will probably restrict the number of cities that can be modeled to one or two. (Airshed modeling for Philadelphia is in progress and the greater New York City metropolitan area is a second candidate.)

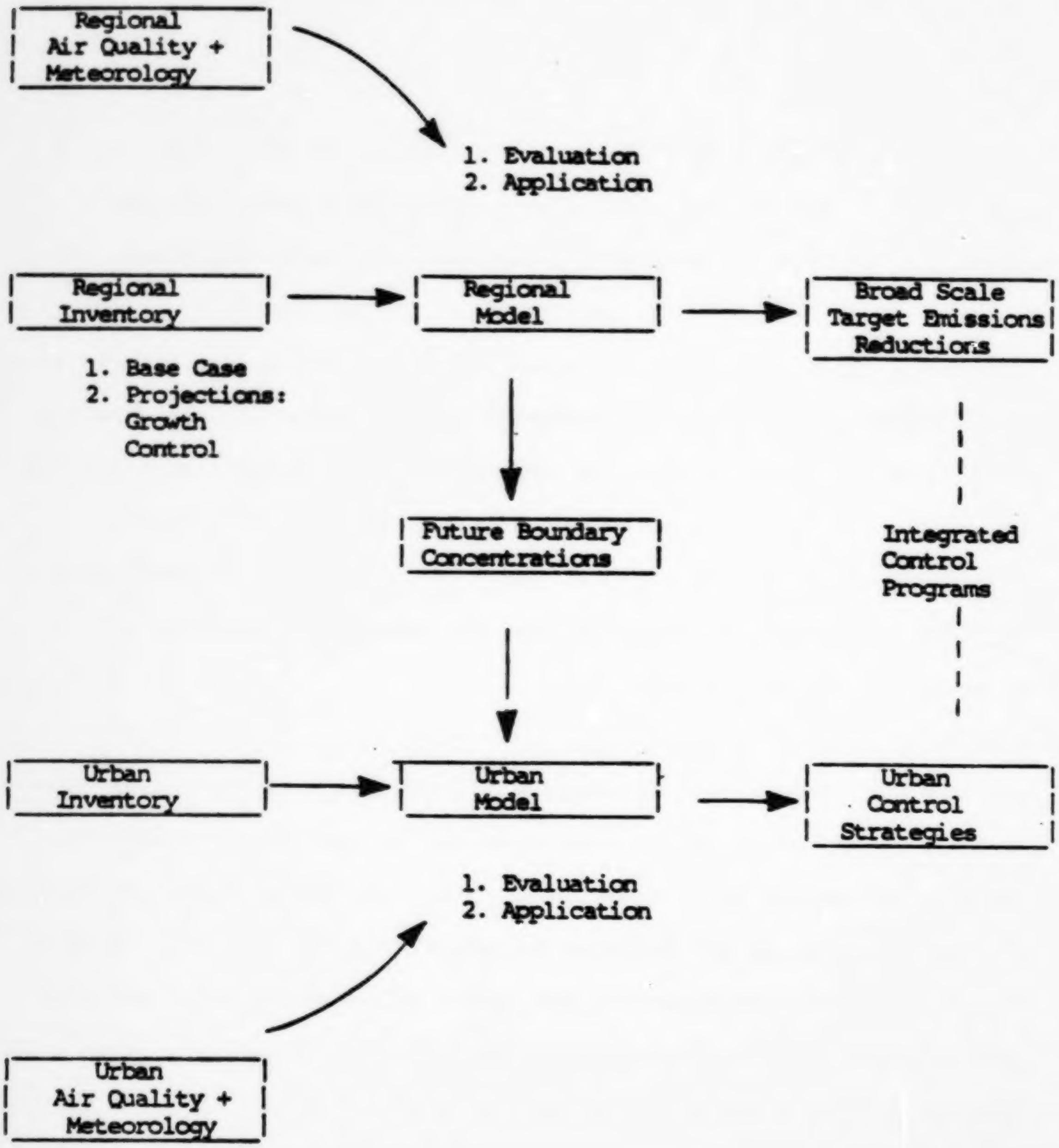
Regional Air Quality + Meteorology
1. Evaluation
2. Application
Regional Inventory
1. Base Case
2. Projections: Growth Control
Regional Model
Broad Scale Target Emissions Reductions
Future Boundary Concentrations
Integrated Control Programs
Urban Inventory
Urban Model
Urban Control Strategies
1. Evaluation
2. Application
Urban Air Quality + Meteorology

Figure 3. Basic NECRMP concept (how regional and urban modeling interface).

STATUS/SCHEDULE OF NECRMP

The status and schedule of NECRMP are summarized in Table 1. All of the data bases required for regional modeling have been completed or are nearly complete. ROM verification activities are scheduled for 1984 and 1985, and regional control tests will be conducted in 1986. With regard to the urban scale, extensive modeling will be conducted for Philadelphia in 1983 and 1984. During this period, the Office of Air Quality Planning and Standards (OAQPS) will: (1) conduct base-case simulations and model evaluation exercises for Philadelphia, (2) conduct a series of sensitivity tests, and (3) run a limited number of future-year projection and control-strategy simulations based on rough estimates of boundary concentrations upwind of Philadelphia. The tests for the latter activity can be rerun by using the boundary concentrations output from ROM for the 1986-1987 time frame.

ORGANIZATIONAL STRUCTURE--EPA/STATE COOPERATION

Table 2 shows the organizational structure of NECRMP. The role of the NECRMP Policy Group is to set the overall direction of the project and is composed largely of the directors of state air pollution control agencies and air branch chiefs of the regional EPA offices. OAQPS and the Office of Research and Development, Environmental Sciences Research Laboratory (ORD/ESRL), are

TABLE 1. SCHEDULE OF NECRMP ACTIVITIES

Status	Activities[a]
Completed	Air quality/meteorological field programs (1979, 1980)--data bases now available.
	Annual county-based emissions inventories--complete spatial, temporal, and species allocation files available; some remaining problems.
	Automated regional inventory data handling system--on-line.
	First generation ROM--on-line.
Remaining	Episode characterization/selection of test days (fall 1983).
	Complete regional emissions inventory for ROM (late 1983)
	Complete second-generation ROM (spring 1984).
	Complete validation of ROM-2 (spring 1985).
	Complete projection and control strategy emission tapes (spring 1985).
	Simulations for projection and control strategies 1986.

[a] All dates are in calendar years.

TABLE 2. NECRMP ORGANIZATION

Group	Composition	Function
Interagency Policy Group	Administrators from States, EPA (regional offices, Office of Air Quality Planning and Standards, and Office of Research and Development)	Overall direction of project, recommendations on priorities, policy questions.
Interagency Model Work Group	Technical representatives of States, EPA (regional offices, Office of Air Quality Planning and Standards, and Office of Research and Development)	Formulation of modeling protocol, resolution of technical issues, recommendations to Policy Group and to EPA.
States, regional EPA offices	----------	Funding/resources for emissions/air quality data bases.
Office of Air Quality Planning and Standards	----------	Overall project coordination/management. Chair Policy and Work Groups. Final emissions inventory. Aerometric analyses (episodes).
Office of Research and Development	----------	Development, refinement, validation, application of ROM.

represented at upper management levels. The NECRMP Work Group, which consists of technical representatives of the states, ORD, and OAQPS, is responsible for developing modeling objectives and procedures that are consistent with the directives of the Policy Group. The interagency composition at both the policy and technical levels is designed to ensure that the modeling protocol will be responsive to the needs and limitations of the participating agencies. The EPA project manager serves as the chairman of the Work Group and the executive secretary (ex officio) of the Policy Group.

EPA's responsibilities are as follows: ORD/ESRL is responsible for developing, testing, and applying the ROM. OAQPS is responsible for coordinating the project, developing the ROM input data bases (emissions, air quality, and meteorological), and conducting the Philadelphia airshed model study. State and local agencies are responsible for supplying the basic emissions inventories and considerable air quality and meteorological data to OAQPS and for participating in planning the program as discussed above.

NECRMP FUNDING AND RESOURCES

Approximately $8 million have been spent thus far on NECRMP (about two-thirds by EPA and one-third by the states), and an additional $4 to $5 million will be needed to complete the project. Major emissions and aerometric data bases have been assembled largely by contractors. Analyses of O_3 episodes and plume transport and transformation have been performed internally and by contractors. Preparation of the final ROM model inputs (emissions, meteorology, and air quality) is being done largely in-house, as are the development, testing, and application of the ROM. The Philadelphia modeling project is being carried out by a contractor under the supervision of OAQPS.

QUESTIONS ADDRESSED IN NECRMP MODELING

The Policy Group and the Work Group are currently identifying the questions to be tested with the ROM (and urban-scale models). Although it is not possible at present to give a definitive list of these questions, examples are given in Table 3.

The general procedure that will be used to answer questions related to control strategies is shown in Table 4. The first step is to identify days that represent the important types of high O_3 episodes in the region. (The premise is that the meteorological conditions of the test days will represent those associated with similar episodes in future years.) The next step is to model the same test days by using "baseline projection" emissions inventories for a future year in which attainment is required. (The baseline projections reflect changes in emissions associated with changes in population and economic activity and also scheduled reductions mandated by control programs already in place.) The results of these model simulations are then used to determine the types of episodes that are likely to require greater control. These episodes will then be modeled again to determine the types of additional control programs that will be required for attainment of the O_3 NAAQS. As stated previously, parallel studies will be conducted with urban-scale models in order to determine the type of control programs that may be necessary to assure compliance in the urban areas and downwind environs. The urban analyses will use forecasts of future-year boundary concentrations supplied by the ROM.

TABLE 3. EXAMPLES OF QUESTIONS TO BE ANSWERED BY THE STUDY

1. What are the relative contributions of transported and urban (local) emissions in different parts of the region? What are the relative contributions of different source regions to high O_3 in different parts of the region?

2. What are the relative contributions of various types of sources to high O_3 occurrences, e.g., stationary sources vs. mobile sources?

3. How will boundary concentrations for urban areas change in the future? (Boundary concentrations are used as inputs for urban-scale photochemical modeling.)

4. What levels of precursor control are required to attain the NAAQS for O_3 and how effective are various types of control programs or approaches? Specific questions related to control:

   a. If the region is approached as a whole, what level of precursor control (% NMHC reduction) is required to attain the NAAQS?

   b. How is the estimate in (a) affected by regional changes in the level of NO_x emissions?

   c. What percent reductions in NMHC (and/or NO_x) are required for the different urban regions in order to reduce regional O_3 values to the NAAQS?

   d. Does it make sense to reduce emissions of sources located in attainment regions?

   e. What is the relative impact (effectiveness) of reducing emissions from mobile vs. stationary sources?

   f. What would be the effect of changes in regional fuel composition or of changes in currently mandated automobile emission control programs?

   g. How effective are specific control measures on a regional basis, e.g., substitution of solvents, vapor recovery measures, traffic reduction?

TABLE 4. BASIC NECRMP APPROACH

1. Identify important types of high O_3 episodes.

2. Perform base-case ROM simulation/model validation.

3. Develop future-year projection emissions inventories.

4. Perform future-year projection simulations with ROM.

5. From the results of (4), determine which episodes/areas require additional control.

6. Prepare various types of "control scenario" emissions inventories.[a]

7. Apply ROM for the critical episodes in (5) by using various control scenario inventories (6).

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[a] Initial simulations will be based on simple sensitivity tests to focus on control requirements. This will be followed by more specific control programs.

MAJOR DIFFICULTIES AND PROBLEMS

Many problems associated with NECRMP are related to the enormous size and complexity of the program. The large number of agencies and tasks involved requires careful coordination on a continuing basis. In addition, circumstances and perceptions are different in each state, and it is not surprising that positions on key items and the degree of interest in the project vary from agency to agency. Furthermore, large spending requirements have come at a time of shrinking budgets. Diminished levels of funding have reduced the ability of the states to assemble urban emissions inventories, and the number of cities for which the Urban Airshed Model can be used has been reduced substantially. Perhaps the greatest problem is the long lead time and uncertainty associated with the project. The possibility remains that major technical (or funding) problems will cause delays that will jeopardize the regulatory utility of the project. Another serious problem is that there is presently no firm legal basis for converting the results of NECRMP analyses into binding multistate air pollution control programs. Despite the problems and uncertainties, the participants have continued to support NECRMP as a viable, and perhaps the best, means for developing controls that are equitable and effective.

CONCLUSIONS

This paper has described EPA's major ROM program, NECRMP. This program was initiated in the late 1970s and is scheduled for completion in 1987. Continued support for this program stems from the need to develop control programs that are effective and fair and that are based on sound scientific analyses. The use of refined state-of-the-art photochemical models that incorporate the transport of O_3 and precursors provides a more credible basis for expensive control programs than do simple models such as EKMA.

A major beneficial result is the joint EPA/state participation in regional modeling efforts. The establishment of the Interagency Policy and Work Groups in 1981 has increased the interest and involvement of the states in NECRMP. The interagency committees have not only contributed to the flow of information but have also been useful in working through complex problems and in resolving the differences between agencies. The cooperative process in molding the study will hopefully lead to multistate control strategies that are perceived as fair and effective. Nevertheless, there are no provisions in the current Clean Air Act Amendments that legally bind the States to use the results of regional modeling efforts. The currently mandated SIP approach is not well suited to the regional nature of the O_3 problem and has led to considerable confusion and conflict. Moreover, regional efforts such as NECRMP, although beneficial, operate on a purely voluntary basis. The answer to this problem may lie in revising the amendments to permit multistate/EPA implementation programs for pollutants and areas that are strongly affected by regional transport.

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NEEDS AND APPLICATIONS OF REGIONAL AIR QUALITY SIMULATION MODELS FOR OXIDANTS IN EUROPE*

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INTRODUCTION

Although the oxidant problem in Europe has not reached the level it has in the United States, it has become a subject of growing concern, especially because of its possible link with acidification. This paper emphasizes the Dutch situation in particular, because we understand the circumstances prevailing in that part of Europe. However, the Dutch situation can often be viewed as representative of larger parts of Western Europe as well.

Although The Netherlands is situated at a high latitude (52° N), substantial O_3 levels can prevail in the summer season. For example, the EPA standard of 120 ppb (240 $\mu g/m^3$) was surpassed on 13 days during the summer of 1982. During such episodes, when easterly winds prevail, the O_3 levels are usually high over all of Western Europe. Maximum values have reached approximately 270 ppb (540 $\mu g/m3$).

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

In The Netherlands, an infrastructure has been developed to provide information for the formulation of air pollution abatement policies. This structure consists of monitoring data, emissions inventories, research programs, modeling, and the Air Quality Monitoring System (AQMS), which plays a central role in integrating and directing the use of these different types of information.

The Netherlands and surrounding countries have a rather dense air quality measuring network. In The Netherlands, an area of 34,000 km^2, hourly concentrations are available for SO_2 (200 stations), NO and NO_2 (92 stations), O_3 (30 stations), and CO (41 stations). This information is supplemented with measurments taken at selected sites, particularly measurements on the determination of compounds of interest to photochemistry. It includes the analysis of up to 200 different chemical species and the physical and chemical structure of aerosols. In addition to ground-level measurements, aircraft measurements are conducted. Flowers and crops are also used as biological indicators of air pollution. Meteorological information, such as wind speed, wind direction, and temperature, is generated by over 60 stations. Higher level information can be obtained from a meteorological mast (200 m) and several other masts. A detailed emissions inventory on a 1 km x 1 km scale is available. Information from this inventory has been processed for model applications, including photochemistry.

To indicate the photochemical patterns of ambient air quality in The Netherlands, Figure 1 shows the O_3 and NO_2 pattern for a 20-yr period. Figure 2 gives general trends for significant air pollutants in The Netherlands.

Although SO_2 levels have decreased during the last two decades, there has been no decrease in photochemically related pollutants. The concentration of PAN, for example, has increased significantly.

DESCRIPTION OF THE DUTCH AIR QUALITY MANAGEMENT SYSTEM

In 1978, the Ministry of Housing, Physical Planning and Environment instructed The Netherlands Organization for Applied Scientific Research (TNO) to construct the Air Quality Management System (AQMS) in an attempt to quantify the diffuse information that eventually leads to policy decisions. The system contains a socio-economic module (based on scenarios for the future), a transmission module, and a module containing consequences accessible to assessment. The interrelations between the modules are of great importance. For AQMS, transport and dispersion models are important tools, but the AQMS also compels modelers to adapt their models to policy purposes. Thus, models should be practical and should not aim at achieving more detail and accuracy than that in accordance with the overall results of the system.

The AQMS for NO_x was recently completed. It contains models for annual averages and higher percentages of NO_2, an episodic SAI airshed model, semi-empirical relations to determine the consequences of NO_x emissions with respect to photochemistry, traffic emission dispersion models, and models for determining wet and dry deposition.

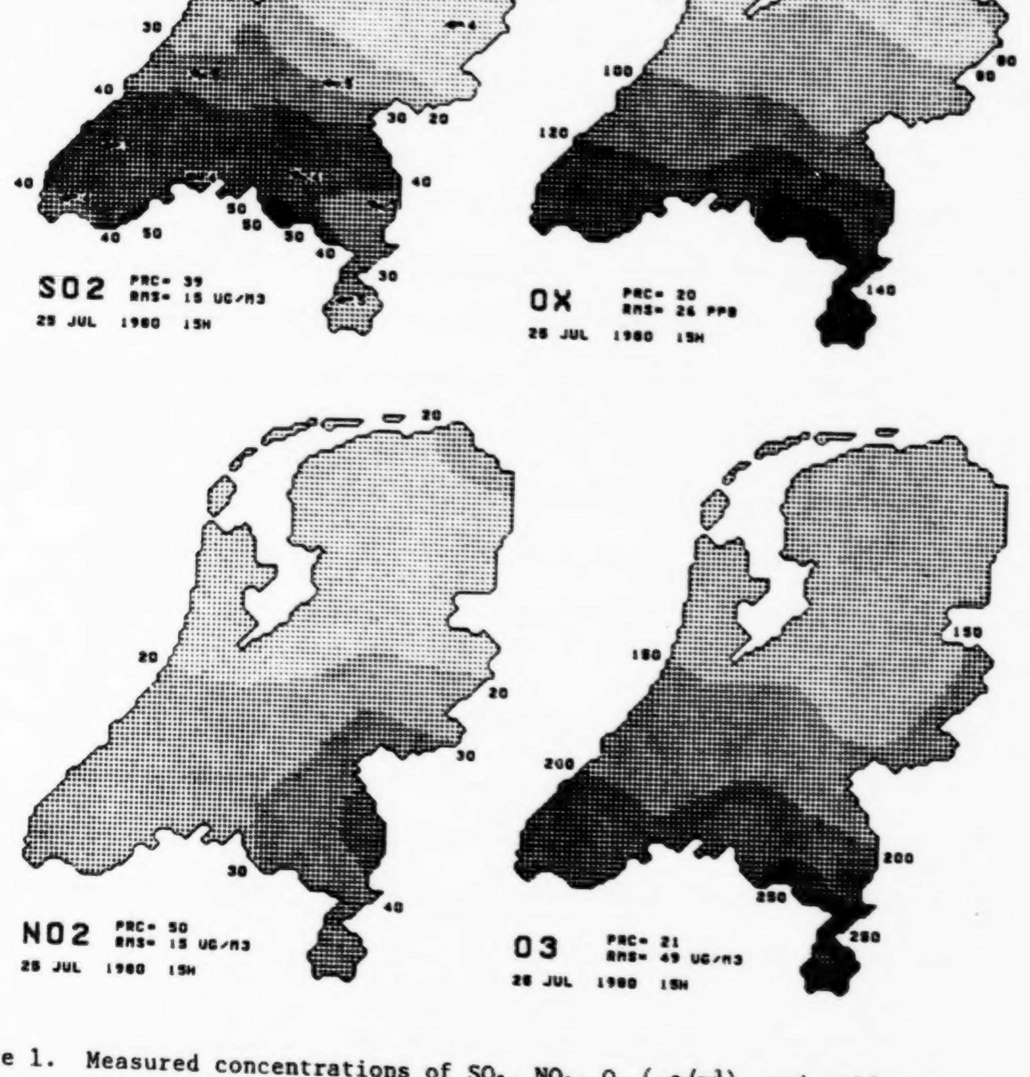
SO2
PRC= 39
RMS= 15 UG/M3
25 JUL 1980 15H
10
20
30
40
50
OX
PRC= 20
RMS= 26 PPB
25 JUL 1980 15H
80
100
120
140
NO2
PRC= 50
RMS= 15 UG/M3
25 JUL 1980 15H
20
30
40
O3
PRC= 21
RMS= 49 UG/M3
25 JUL 1980 15H
150
200
250

Figure 1. Measured concentrations of SO_2, NO_2, O_3 ($\mu g/m^3$), and oxidants $NO_2 + O_3$ (ppb) for July 25, 1980, 1500 h (Automated Air Quality Monitoring System, Dutch National Institute of Public Health). The arrows indicate windspeed in meters per second.

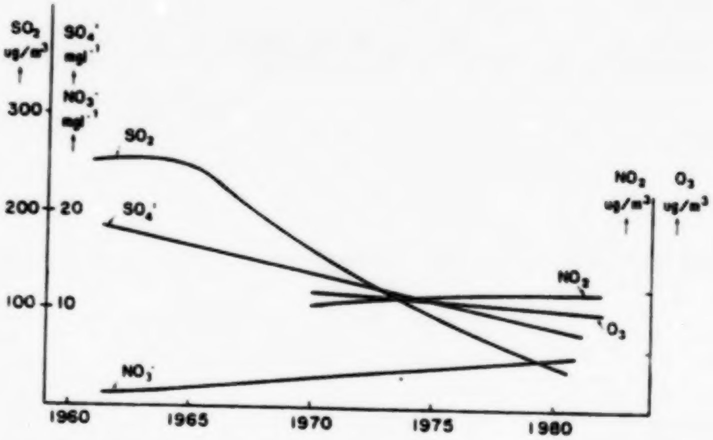

Figure 2. Trend of yearly averaged concentration levels in The Netherlands. $SO_4^=$ and NO_3^- measured in rain water.

Table 1 indicates how the AQMS integrates modeling results and presents them in a manner compatible with policymaking. For a more complete description of the AQMS, see Zwerver (1982) and Bovenkerk et al. (1982).

AIR QUALITY POLICY REQUIREMENTS

The relative position of models in the field of air pollution research may be indicated by the amount of money spent on a model's development and application on the one hand and its infrastructure on the other. Table 2 gives a rough estimate of the money spent by the Ministry over the last 10 yr.

Table 2 suggests that the development and use of models is relativley cheap. However, models require information infrastructures. Although the development of complex models may be relatively cheap, the application costs may be considerable, a factor arguing for relatively simple and practical models.

TABLE 1. THE NO$_x$-AQMS AND ITS RESULTS

Environmental Issue	Standard Violation	Minimal Required Improvement (%)	Minimal Required NO$_x$ reduction (%)
Exposure of humans to NO concentrations	No violation	-	-
Exposure of humans to NO$_x$ concentrations	Close to process industry	50	Optimal
	In cities	5	Dutch traffic, 10
	At street level	25-35	Dutch traffic, 50-70
	In cars	30-35	Dutch traffic, 50-70
	At home	up to 80	regulations for burners, etc.
Exposure of human HNO_3 concentration	No violation	-	-
Visibility	No standard	Improvement, 50-80 20-30	Europe & Netherlands 100, 50
Exposure of human to O_3 concentrations	In over half the country, several days a year	30	Europe & Netherlands NO$_x$, more than 80; HC, more than 40
Exposure of humans to PAN and other photochemical products	No standard	-	-
Exposure of cultivated plants to NO_2	In more than half the country	50-80	Europe & Netherlands, 80
Exposure of cultivated plants to O_3	In more than half the country for several days a year	40	Europe & Netherlands, NO$_x$, 80; HC, 40
Exposure of cultivated plants to PAN	No standard	-	-
Exposure of natural vegetation to NO_2 and O_3	No standard available, but more severe than on cultivated plants for sensible locations	30-40	Europe & Netherlands, 50-80
Exposure of flora, fauna, and ecosystems to acidification		75	Europe & Netherlands. NO$_x$ + SO_2 + NH_3: 75
Exposure to eutrification	No standard	-	-
Exposure of materials to NO_2, O_3, (SO_2) in musea, etc.	Yes	?	Does not apply
Exposure of materials to NO_2, nitrates, SO_2, sulphates	No standard	To avoid any damage	Europe & Netherlands, NO$_x$ 70, SO_2 50
Climatological changes	No standard	-	-

TABLE 2. ESTIMATED BREAKDOWN OF MONEY SPENT BY MINISTRY OF HOUSING, PHYSICAL PLANNING AND ENVIRONMENT

Project	Expenditure (million Dfl.)
Emissions inventory	50
Monitoring network	130
Research on photochemistry	15
Model development and application	15
AQMS structure	7

Requirements Related to Resolution and Degree of Discrimination

Questions regarding resolution in time and space, the degree of discrimination, the abatement of traffic and industry emissions, and the efficiency of NO_x and/or HC abatement arise in model applications. At the moment, policymakers need rather simple answers to these questions, although the questions themselves probably cannot be answered easily. The problems that arise are: What are the essential details and how sophisticated should the model be in order to determine which compounds should be abated, what spatal scale of abatement is required, and what are the quantitative results.

Ozone formation is a large-scale phenomenon, and O_3 concentrations show a rather flat pattern. So, model results should indicate the minimum spatial dimensions of the areas to which abatement should be applied and the compounds that should be abated (HC, NO_x, or both). The fact that O_3 formation is not a linear process implies that several abatement strategies should be separately calculated in order find the most efficient one. Then, the problem of O_3 variance and abatement on a smaller scale becomes apparent, and large-scale models can be used to define the boundary conditions for smaller areas (including individual plumes). As stated previously, the oxidant problem is just one of many air quality problems facing The Netherlands. In Table 3, the authors give their personal views of present environmental priorities.

APPLICATION OF PHOTOCHEMICAL DISPERSION MODELS IN THE NETHERLANDS

History

Photochemical phenomena have been investigated in The Netherlands since approximately 1970. This field of investigation was soon extended from the development of measuring devices. In addition to conducting smog chamber experiments and making detailed analyses of the phenomena, researchers developed and applied models to practical situations. First, the EKMA approach and box models were used; later, the SAI airshed model was used. Much more detailed field experiments were also carried out, including the use of airplanes and the measurement of over a hundred different HCs. Table 4 gives an overview of the historical development of photochemical dispersion models (see also Guicherit et al., 1978).

TABLE 3. PRIORITY AND MODEL PERFORMANCE RELATED TO ENVIRONMENTAL ASPECTS AND ESTIMATED NEEDS FOR ABATEMENT[a]

Environmental Aspects, Concentrations, Phenomena, and Subjects	Spatial	Scale and Resolution Time	Grid	Priority
O_3 concentrations				
Humans	Europe	Episodic (days)	$\sim$20 x 20 km^2	1-2
Crops	Europe	Episodic (days)	$\sim$20 x 20 km^2	3
Natural vegetation	Europe	Episodic (days)	$\sim$20 x 20 km^2	3 (1 if O_3 is main cause cause of dying forests)
NO_2 concentrations				
Humans	Mesoscale	98th percentile	1-400 km^2	2-3
Crops	Mesoscale	Hourly based	1-400 km^2	3
Natural vegetation	Mesoscale	Hourly based	1-400 km^2	3
Humans in houses, cars, and traffic	Local and microscale	Hourly peaks	Population exposure statistics	1-2 (1 if O_3 is also strongly involved; subject of Dutch inhalation toxicology research
Acidification	Europe	Long-term[b]	Depends on land use, 400 km^2 up to the size of country	1
Eutrification	National scale (NH_3)	Long-term		1
Dying forests		Long-term		1
Secondary products (PAN, HNO_3, aerosols carcinogenics aldehydes, etc.)	Mesoscale	98th percentile Hourly based[c]	1-400 km^2	1-3?
Visibility	Mesoscale	Episodic	Vertical columns, countries	3
Climate	From continental to microscale	Long-term, episodic	-	1-3?
Materials	Europe/ mesoscale	Long-term	400 km^2, countries	3

[a] See Table 1 for preliminary estimates of necessary abatement.

[b] Outside The Netherlands (e.g., Scandinavia), the problem has a more or less episodic character (episodic rains). In The Netherlands, dry deposition contributes most in the long term.

[c] For deposition, long-term and 400 km^2 up to size of country.

TABLE 4. HISTORICAL OVERVIEW OF DUTCH RESEARCH ON PHOTOCHEMISTRY

Year	Research Activity	Major Emphasis of Activity
1970	Development of monitoring methods and instruments (O_3, NO_x, HC, UV, PAN)	Monitoring equipment
1975	Monitoring of mainly gaseous components	Intrusion of O_3
	Natural O_3 and PAN levels	Urban scale
	Trend analysis	Ratio of contributions of traffic and industry
	Models (Box model, EKMA variant)	HC/NO_x abatement
	Smog chamber research	
	Rough abatement scenarios	
1978	Integrated research programs:	
	Monitoring & physical characterization of aerosols, gaseous components	Contribution of natural emissions
	Smog chamber research of aromatics	
	"Aged smog" (sampled at 200 m)	Relative importance of the pollutant inflow
	Conversion of SO_2 interacting with aerosols	
	Monitoring method for OH radicals (failed)	Traffic/industry
	Reactive NMHC detector	HC/NO_x abatement
	Preparation for and application of models (EKMA and SAI airshed models)	
1982	Mesoscale and large-scale models	
	Plume modeling	

The EKMA Approach

An EKMA-type approach, suited especially to the Dutch situation and developed by Guicherit (1978), has been used to gain an initial insight into photochemical processes. The EKMA approach has also been used in the framework of a large project, aimed at the experimental determination of oxidant levels in The Netherlands. Measurements were performed at a 200-m-high meteorological mast, and several flights were conducted. The EKMA approach is used to determine, in a qualitative manner, the influence of Dutch sources on O_3 formation. The results indicate that this influence is relatively small. Combining the results of aircraft measurements (see Figure 3) and EKMA calculations led to rough estimates of downwind contributions to the O_3 levels (see Table 5) of typical emitters. At the moment, an EKMA-type box model is used in a trajectory mode in combination with the SAI airshed model. A few runs are being conducted with the airshed model, supplemented with box model trajectory calculations, in which both EKMA and other chemical mechanisms are incorporated.

The Oxidant Approach

An analysis of NO, NO_2, and O_3 data from the Dutch National Monitoring Network showed the limiting influence of O_x (= $NO_2 + O_3$) levels on NO_2 formation, indicating a possible important inflow of O_x, O_3, or O_3 precursors in our area. These oxidant levels were mainly determined by O_3 inflow across the Dutch borders. In favor of the Dutch Environmental Council, which is preparing its comments on the NO_2 standard proposed by the Dutch Council for Public

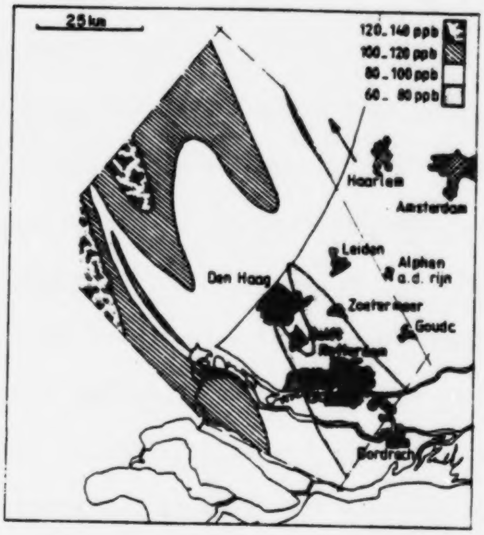
25 km
120 - 140 ppb
100 - 120 ppb
80 - 100 ppb
60 - 80 ppb
Haarlem
Amsterdam
Leiden
Alphen a.d. rijn
Den Haag
Zoetermeer
Gouda
Rotterdam
Dordrecht

Figure 3. Example of measured O_3 profiles at 650-m height above Rijnmond area.

TABLE 5. DOWNWIND CONTRIBUTIONS TO O_3 LEVEL

Emitter/Pollution Source	Approximate Increase of O_3 Downwind from Source (ppb)
City (100,000 inhabitants)	1
Rijnmond (Rotterdam industrial area)	20
Southern part of The Netherlands	10
Transboundary inflow of precursors	20
Transboundary inflow of O_3	80

Health, Van Egmond et al. (1982) developed and applied a simple oxidant model to establish a simple, empirical relation between the probability of exceeding the NO_2 standard and the NO_x concentration levels (see Figure 4). This simplified

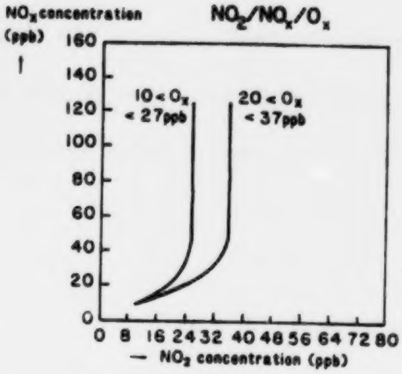

Figure 4. Limitation of the NO_2 level (limit depends on the actual NO_x and oxidant levels).

relationship, based on the photostationary equilibrium of NO, NO_2, and O_3, has been applied in a three-layer numerical grid model (Van Egmond, 1982(b)).

The model, incorporating a very simple chemical scheme, accurately described the O_x and NO_2 situation in The Netherlands, once again indicating the large inflow of oxidants into the Dutch area.

Nonepisodic Modeling of NO_2 Concentrations

Although not directly related to O_3 levels, the influence of the inflow of primary and secondary pollutants on a long-term basis is nevertheless worth mentioning. In terms of NO_2 frequency distributions and estimates of yearly nitrate depositions in Dutch territory, van den Hout et al. (1983) applied a Gaussian-type model combined with wind-direction-dependent empirical factors for the NO_2/NO_x conversion. This model, combined with other approaches (the oxidant model and a preliminary microscale model), has led to the results presented in

Table 2. These results also demonstrate the importance of inflow from outside The Netherlands. Similar considerations apply to the deposition of nitrates. About 70% of this deposition originates from across the border and contributes 20% to the acidification of the Dutch soil. Vice versa, Dutch NO_x emissions are deposited in other countries.

Application of the SAI Airshed Model

SAI airshed model calculations were carried out for an episode of relatively high oxidant levels (June 7-8, 1976). Runs to test the model and establish its sensitivity to the assumed boundary conditions were followed by runs for application purposes. In these runs, several abatement reference cases involving traffic and industry NO_x and HC emissions were simulated.

Figure 5 shows the region covered by the model (310 km x 230 km). In this region, three large industrial areas are situated at distances from each other of 100 m up to 300 km. These are the Rijnmond (Rotterdam harbor) area in the Western part of The Netherlands, the Antwerp area in the Western part of Belgium, and the Ruhr area in the Western part of Germany. In addition to the industrial and urban areas, the region covered by the model includes rural areas with low NO_x and HC emission densities.

The well-known SAI airshed model (Reynolds, 1979) was originally developed for urban-scale photochemical episodes. For this application, the model has been "stretched out" in space to regional scale. Hourly concentrations of several air pollutants, O_3, PAN, NO, NO_2, and four HC classes have been

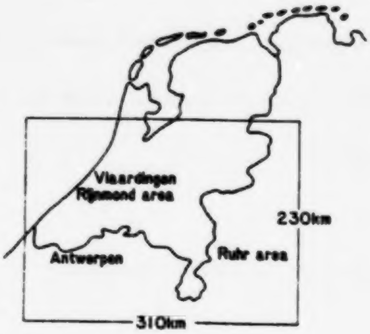

Figure 5. The modeling area.

calculated for 10-km grid distances; five levels have been used in the vertical direction.

Test Runs--

The amount of pollutant inflow into the area is uncertain. For this reason, boundary conditions were changed in order to test the sensitivity of the model and to check how realistic the assumed boundary conditions were.

The originally selected conditions gave the best overall agreement with the monitoring data. Generally, the results of the calculations fit and correlated quite well with the measurement data. For instance, a calculated maximum O_3 value of 244 $\mu g/m^3$, averaged over 11 monitoring stations, compares very well with a measurement of 259 $\mu g/m^3$. The temporal correlation coefficient for monitoring stations with O_3 values higher than 40 $\mu g/m^3$ was 0.91; the spatial correlation of the maxima was 0.62.

Figure 6 shows observed and calculated measurements (O_3, NO and NO_2) for a monitoring site in the industrial Rijnmond area (for further information see Builtjes et al., 1980, 1982).

Application of the Model in the Establishment of an NO_x Policy--

The main issue facing Dutch policymakers attempting to counteract the effect of pollutants with a long lifetime is: How effective is domestic abatement in a small country like ours in comparison with abatement measures taken abroad? How do the main sources (traffic and industry) contribute to environmental pollution inside and outside the country? The answers to these questions are complicated by the nonlinear behavior of photochemical transformations; consequently, it is not possible to express the pure and independent contribution of one source to the concentrations of O_3 and NO_2 without considering the emissions from other sources.

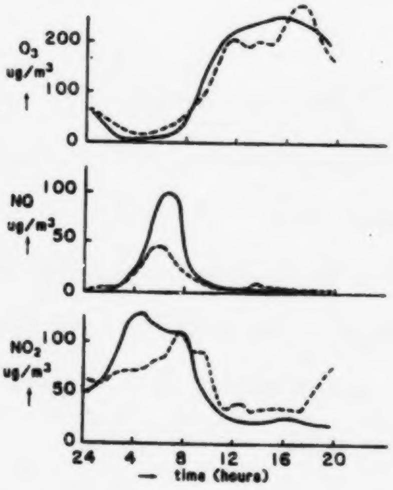

Figure 6. Concentrations at Viaardingen. Dashed line represents observed concentrations; solid line represents calculated concentrations.

In order to gain an insight into the effectiveness of emission reduction, two sets of runs were carried out, directed to source categories and to species. The results of the first set are shown in Figure 7 and refer to the influence of a 100% reduction of NO_x and HC emissions from traffic and industry. The O_3 concentrations were nearly unaffected in all cases. Even with a 100% reduction of all emissions in the area, 85% of the O_3 still remains; the efficiency with respect to NO_2 concentrations is also relatively low, but a 100% reduction of NO_x and HC emissions finally drops the NO_2 concentrations to a a low level.

The results of the second set of runs, shown in Table 6, show separate and combined reductions of NO_x and HC emissions in the considered area of 0%, 40%, and 100%. It is apparent that a 40% reduction of the HC emission yields a decrease in the maximum O_3 emission by 2%. This effect is small, partly because the flux of background HC across the boundaries into the model region is much

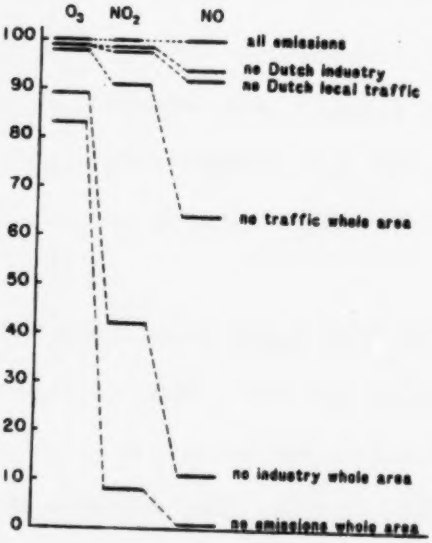

Figure 7. Average reduction of maximum hourly O_3, NO_2, and NO concentrations.

TABLE 6. EFFECT OF EMISSION REDUCTIONS ON THE CALCULATED MAXIMUM O_3[a]

Run	Emission Reduction in Model Region (Species)	(%)	Maximum O_3 ($\mu g/m^3$)
A	None		275
B	HC	40	269
C	HC	100	250
D	NO_x	40	274
E	NO_x	100	205
F	HC and NO_x	100	206

[a] HC emission is 105,000 kg/h; NO_x emission is 195,000 kg/h.

the flux of background HC across the boundaries into the model region is much larger than the HC emission inside the region. For NO_x, on the other hand, the background flux across the boundaries is smaller than the emission inside the region. Consequently, for a larger area the effect of a 40% NO_x emission reduction on O_3 concentrations will be much smaller than that of a 40% HC emission reduction, a fact that argues for HC control.

An important result of these model calculations is that a reduction in emissions in the model region has only a limited effect on the O_3 maxima. The influence of the pollutants from sources upwind of the region is much more important than that of local sources. The estimates of the upwind concentrations resulting from remote sources are very uncertain, however. The HC/NO_x ratio in this background air, which is important for O_3 formation efficiency, can be much larger than the HC/NO_x emission ratio of the model the region. For the development of efficient control strategies for situations like the one discussed here, it is therefore necessary to perform calculations on a spatial scale that is considerably larger than the 300-km scale used in these calculations (see also Van den Hout et al., 1982).

In view of the above results, it should be clear that The Netherlands, to develop abatement strategies in their own country and to stimulate international actions, would welcome research on and clarification of O_3 and O_x (= O_3 + NO_2) formation on a large scale, including the use and/or development of large-scale models.

One problem needing careful consideration is the relative efficiency of NO_x and HC abatement, in particular the emissions produced by motor vehicles. At the moment, discussions are going on in the European Community about a further reduction of CO and HC traffic emissions. The discussions include a detailed consideration of matters like the European test cycle. However, the definition of this cycle, the resulting traffic regulations, and future developments in the construction of motor cars will determine the admissible levels of HC and NO_x car emissions and the ratio between the two in the coming 20 yr. So, a simple or complicated model that can discriminate between abatement efficiences for NO_x and HC on a European scale would fit the European needs very well.

The tight time schedule for the above-mentioned activities favors the use of directly applicable models instead of waiting for more complicated models that need further development and testing or collecting detailed input data.

In conclusion, it is still unclear whether an unambiguous solution can be found for such phenomena as the ones discussed here. Different chemical submodels lead to different estimates of what would be the most favorable solution for the Dutch situation in so far as substantial NO_x and/or HC emission reduction on a European scale is considered. The margin of uncertainty seems to be too wide for reliable interpretation. Table 7 demonstrates this uncertainty. It gives estimates from several Dutch experts, based on their model calculations and experience with respect to the possible influence of NO_x and HC emission reduction for O_3 levels on a European scale. These are very speculative estimates, given here only to illustrate the dilemma and the need for large-scale models.

CONCLUSIONS AND REMARKS

The application of models in The Netherlands indicates that O_3 formation is due to large-scale phenomena caused by NO_x and HC emissions on a European scale. In order to be effective, abatement strategies should be developed with the continental scale in mind. Large-scale models applied to a substantial part of Western Europe, at least on a scale of a 1,000 km x 1000 km, may contribute to the establishment of these abatement strategies. One major problem is the effectiveness of NO_x and/or HC control on the European scale. Currently used chemical submodels give different answers to this problem. Clarification of this point could also contribute to the discussions being held in the European Community's working group, ERGA, with respect to the abatement of traffic emissions. In addition to O_3, other compounds and phenomena must be considered

TABLE 7. ESTIMATED O_3 REDUCTION ON A EUROPEAN SCALE AS A CONSEQUENCE OF HC AND/OR NO_x EMISSION REDUCTION

NO_x Reduction (%)	HC Reduction (%)	O_3 Reduction (%)
30	0	0
60	0	15
90	0	50
0	15	10?
0	30	25?
0	60	50?
30	15	10-15??
60	30	20-40??
60	60	20-60??

on a mesoscale and a large scale, especially the problems related to acidification.

ACKNOWLEDGMENTS

This paper gives an overview of photochemistry work that was conducted by experts in the field, to whom the authors are greatly indebted.

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DISCUSSION

J. Shreffler: There is a very strong modeling and measurement program in The Netherlands, as seen from this presentation. One of the major isues to be addressed at the conference should be: If we go to regional modeling, who will do it an how will it be done to put together a package of emissions data, aerometric data, for the large number of European countries? Among the countries, are the measurement equipment and procedures the same? In other words, how do we get a compatible data base for a large number of countries?

APPENDIX A. GUIDELINES FOR EMISSIONS INVENTORY PRESENTATIONS

Data base name/source	General Emission Inventory in The Netherlands
Reasons for inventory development?	Air and water quality management
Who collects the raw data? (private industry, national/provincial government, etc.)	Dutch Organization for Applied Research (TNO) by Order of the Ministry of Pub. Health and Env. Protection and the Ministry of Traffic and Public Works
How is raw data collected? (questionnaire, permit system, inspection, other)	Large industries: inspection Small industries: questionnaire exc. those of significant env. concern; also inspection
How frequent are data updated?	Combustion sources >20 MWh, yearly; all other sources, once in 3 yr.
Are updates legally required?	No
List legal or confidentiality restrictions which may prevent release of the data	Emis. data from private industry are confidential; limited clustered data may be published
Area of coverage	Netherlands (ca. 40,000 km^2)
Coordinate system	Dutch topographical map
Point source information; define a point source	Vertical stacks and chimneys
A) Raw data collected:	
List stack information	Locn. (10m), height (m), cross sect. area (mm^2), name
List major contributing source categories (industries)	Food, paper, refining, chems., bldg. mat'l., prim. Metal, metal products, thermal generation, coke, other industry
List types of raw data collected and temporal resolution where appropriate	See Appendix C.
Spatial resolution	10 m
Dates of available data	Previous yr. with regard to yr. of registration
B) Emission estimates:	
List pollutant species	1500 subtances (air and water) (Appendix B)
Temporal resolution of calculated emissions	See Appendix C.
List information available for temporal apportionment	Information obtained from plant officials
List percentage of emissions estimated by following methods:	
_____ Standard emission factors with specific plant information	15
_____ Nonstandard emission factors with specific plant information	63
_____ Source test	} process emissions only
_____ Material balance	18
_____ Other, specify	4
What emission factors, if any, are used?	
List publication describing emission factor development program	See Handbook of Emission Factors, Part 2, Industrial Sources (In press).

Are reported emission controlled or uncontrolled?	Both
Are control equipment and efficiency information available?	yes
Describe method of estimating volatile organic compound emissions	See Handbook of Emission Factors, Part 1, Nonindustrial sources.
Area source information; define an area source	Bldgs. with openings; open areas (e.g., storage of petroleum liquids; chem. plant; Ore and coal handling
A) Raw data collected	
List major contributing source categories	
List subclasses of stationary area mobile sources	
List types of raw data collected, spatial and temporal resolution where appropriate	See Appendix C.
Dates of available data	Previous yr. with regard to yr. of registration
B) Emission estimates:	
List pollutant species	See Appendix B.
Temporal resolution of calculated emissions	See Appendix C.
List information available for temporal apportionment	Information obtained from plant officials
Describe grid system or spatial resolution	Coordinates of mid-point of terrain
List information available for spatial approtionment	
Are published standard procedures used for temporal and spatial allocation and emission calculations?	No
If yes, list major references	
Describe method of estimating volatile organic compound	See Appendix B.
General emissions	
Comment on completeness	As complete as possible within the scope of the project
Comment on currentness	First round, 1974-1981 Second round, 1982-1984 Combustion sources >20 MWh, yearly
Summarize Quality Assurance Program	Forms (see Appendix C) are inspected. After discussion with relevant registrator and approval, a decoded printout is made and again inspected. After final approval, decoded printout is sent to plant officials who may comment within 3 mo.

Who is responsible for data quality? TNO

Are source inventory data handled manually or by computer? By computer

Attach detailed record formats

APPENDIX B. LIST OF POLLUTANT SPECIES

2nd round (1982-1984)   1st round (1974-1981)

```
EMISSI - CONVERSIELIJST PER NIEUWE STOF
 111 WATER,WATERDAMP                          100.0     133 46 WATER

1011 METHAAN                                  100.0     139 50 METHAAN

1015 ETHAAN                                   100.0     140 50 ETHAAN
                                              100.0     461 55 KOOLWATERSTOFFEN C2, NNB
1019 PROPAAN                                  100.0      28 50 PROPAAN
                                              100.0      54 55 KOOLWATERSTOFFEN C3, NNB
1023 BUTAAN,N                                 100.0      56 55 KOOLWATERSTOFFEN C4, NNB
                                              100.0     249 52 BUTAAN, N-

1027 ISOBUTAAN                                100.0     151 52 ISOBUTAAN

1031 PENTANEN                                 100.0     148 52 PENTAAN, N-
                                              100.0     574 52 ISOPENTAAN (2-METHYLBUTAAN)
                                              100.0     711 52 KOOLWATERSTOFFEN, ALIFATISCH, C5
1035 HEXAAN,N                                 100.0     150 52 HEXAAN, N-

1039 HEXANEN                                  100.0     734 55 KOOLWATERSTOFFEN C5-C6, NNB
                                              100.0    1633 52 DIMETHYLBUTAAN, 2,2-
1043 HEPTANEN                                 100.0     679 52 HEPTAAN, N-

1047 CYCLOHEXAAN                              100.0      58 53 CYCLOHEXAAN

1051 KWST.,ALIFATISCH,MENGSEL,C2-C10          100.0      52 55 OLIE, FRACTIES KOOKPUNT 100-200  C
                                              100.0     134 99 PETROLEUMETHER-EXTRAHEERBAAR
                                              100.0     135 55 KOOLWATERSTOFFEN, NNB
                                              100.0 1   184 55 KOOLWATERSTOFFEN, VERBR. HOUT
                                              100.0     191 55 NAFTA, KOOKTRAJECT 40-90  C
                                              100.0     193 55 NAFTA, KOOKTRAJECT 40-185  C
                                              100.0     229 55 PETROLEUMETHER (KOOKP.BENZ.) 90-120
                                              100.0     234 55 PETROLEUMETHER (KOOKP.BENZ.)145-164
                                              100.0     340 55 PETROLEUMETHER (KOOKP.BENZ.) 80-110
                                              100.0     358 55 PETROLEUMETHER (KOOKP.BENZ.)100-140
                                              100.0     377 55 PETROLEUMETHER (KOOKP.BENZ.) 55-75
                                              100.0     543 52 OCTAAN, N-
                                              100.0     550 52 KOOLWATERSTOFFEN, ALIF., VERZ., >C3
                                              100.0     583 55 PETROLEUMETHER (KOOKP.BENZ.) 40-60
                                              100.0     589 96 OPLOSMIDDELEN, ORGANISCH, NNB
                                              100.0     678 55 OPLOSMIDDELEN, KOOLWATERSTOFFEN,NNB
                                              100.0     680 52 NONAAN, N-
                                              100.0     712 52 KOOLWATERSTOFFEN, ALIFATISCH, C9
                                              100.0    1081 55 PETROLEUMETHER (KOOKP.BENZ.) 60-95
                                              100.0    1094 96 OPLOSMIDDELEN,ALIF/AROM/GECHL-KWST.
                                              100.0    1191 55 PETROLEUMETHER (KOOKP.BENZ.) 40-135
                                              100.0    1198 55 KOOLWATERSTOFFEN, ALIFATISCH, NNB
                                              100.0    1200 55 PETROLEUMETHER (KOOKP.BENZ.) NNB
                                              100.0    1312 55 KOOLWATERSTOFFEN, BER. ALS METHAAN
                                              100.0    1352 55 KOOLWATERSTOFFEN C1 T/M C5, NNB
                                              100.0    1405 55 KOOLWATERSTOFFEN,KOOKPUNT 140-180 C
                                              100.0    1715 97 KOOLWATERSTOFFEN, ALIF.VERZ.,TOTAAL
                                              100.0    1781 55 KOOLWATERSTOFFEN ALIFATISCH
```

APPENDIX C. EMISSIEREGISTRATIE

Vertrouwelijk. Alleen voor personeel aangewezen
volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Bedrijvenbestand *Plant file*

1- 5	1. Recordcode	3 9 0 9 0	
6	2. Mutatiesoort (code 1)	☐	
7- 12	3. Bedrijfsnummer	☐☐☐☐☐☐	*plant number*
13- 42	4. Naam *plant name*	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
43- 67	5. Vestigingsadres *address*	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
68- 92	6. Vestigingsplaats *town*	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
93- 95	7. Gemeente (code 2)	☐☐☐	*municipality code*
96- 98	8. Ligging: hor. km	☐☐☐	
99-100	*location* 10 m	☐☐	
101-103	ver. km	☐☐☐	
104-105	10 m	☐☐	
106	9. Toezicht lucht (code 3)	☐	*has to do with air quality legislation*
107	10. Toezicht water (code 4)	☐	*" " " " water " "*
108-112	11. Aantal werknemers	☐☐☐☐☐	*number of employees*
	12. Aantal installaties:		
113-114	Totaal	☐☐	*number of (emitting!) installations*
115-116	Te herregistreren	☐☐	
117-118	Aantal grote vuurhaarden	☐☐	*number of large (>20MW$_{th}$) Combustion sources*
119	13. Registratiesoort(code 5)	☐	
120-121	14. Basisjaar	☐☐	*base year*
122-123	15. Opname-instantie(code 6)	☐☐	
124-125	16. Opname-persoon	☐☐	*code of inventory employee*

APPENDIX C. continued

Vertrouwelijk. Alleen voor personeel aangewezen
volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Adressenbestand Address-file

1- 5	1. Recordcode	3 9 0 9 1	
6	2. Mutatiesoort (code 1)	☐	
7- 12	3. Bedrijfsnummer	☐☐☐☐☐☐	plant number
13- 42	4. Naam	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	plant name
43- 67	5. Postadres	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	address
68- 73	6. Postcode	☐☐☐☐ ☐☐	
74- 98	7. Plaatsnaam	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	name of town
99-113	8. Contactpersoon	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	(name of (principal) liaison employee)
114-125	9. Telefoonnummer	☐☐☐☐☐☐☐☐☐☐☐☐	

APPENDIX C. continued

Vertrouwelijk. Alleen voor personeel aangewezen volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Installatiebestand *Installation file*

1- 5	1. Recordcode	39092	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	plant number
13-15	4. Installatienummer	☐☐☐	installation number
16-17	5. Aantal samengenomen	☐☐	number of similar installations
18-21	6. Installatiesoort (code 8)	☐☐☐☐	installation category code
22-25	7. Bedrijfstak (code 7)	☐☐☐☐	industry category code
26-55	8. Naam installation name	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
	9. Ontwerpcapaciteit		
56-59	mantisse	☐☐☐☐	designcapacity
60	exponent	☐	
61	eenheid (code 10)	☐	capacity unit code
62-64	10. Bezettingsgraad (%)	☐☐☐	production level
65-68	11. Bedrijfsuren per jaar	☐☐☐☐	operational hours / year
69-70	12. Bouwjaar	☐☐	year of erection
71-72	13. Verwachte technische levensduur (jaren)	☐☐	technical life-expectation
73-75	14. Produktiewerknemers (aantal)	☐☐☐	number of production-employees
76	15. Inrichtingenbesluit (code 29)	☐	has to do with legislation
77-79	16. Ligging: hor. km	☐☐☐	location
80-81	10 m	☐☐	
82-84	ver. km	☐☐☐	
85-86	10 m	☐☐	
87-88	17. Basisjaar.	☐☐	
89-90	18. Opname-instantie (code 6)	☐☐	
91-92	19. Opname-persoon	☐☐	

APPENDIX C. continued

> Vertrouwelijk. Alleen voor personeel aangewezen volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Apparatenbestand — Apparatus file

1- 5	1. Recordcode	3 9 0 9 3	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	plant number
13-15	4. Installatienummer	☐☐☐	installation number
16-17	5. Apparaatnummer	☐☐	apparatus number
18-20	6. Aantal samengenomen	☐☐☐	number of similar apparatuses
21-24	7. Apparaatsoort (code 9)	☐☐☐☐	apparatus category code
25-54	8. Naam apparatus name	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
	9. Ontwerpcapaciteit		design capacity
55-58	mantisse	☐☐☐☐	
59	exponent	☐	
60	eenheid (code 10)	☐	capacity unit code
	10. Brandstofverbruik		fuel consumption
61-62	soort (code 11)	☐☐	fuel code
63-66	mantisse	☐☐☐☐	
67	exponent	☐	
68	eenheid (code 12)	☐	consumption unit code
69-70	11. Stookwijze (code 27) Brandersituering (code 28)	☐☐	combustion technique code + burner geometry code
71-72	12. Basisjaar	☐☐	
73-74	13. Opname-instantie (code 6)	☐☐	
75-76	14. Opname-persoon	☐☐	

APPENDIX C. continued

Vertrouwelijk. Alleen voor personeel aangewezen volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Bronnenbestand lucht Source file (air)

1- 5	1. Recordcode	3 9 0 9 4	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	plant number
13-15	4. Bronnummer	☐☐☐	Source number
16-17	5. Aantal emissiepunten	☐☐	number of places where emissions occur
18	6. Bronsoort (code 13)	☐	Source category code
19-38	7. Naam Source name	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
39-41	8. Ligging: hor. km	☐☐☐	
42-43	10 m	☐☐	location
44-46	ver. km	☐☐☐	
47-48	10 m	☐☐	
49-51	9. Gemeente (code 2)	☐☐☐	municipality code
52-54	10. Geod. hoogte (m) + 10 m	☐☐☐	
55-57	11. Geom. hoogte	☐☐☐	height of source
	12. Bronoppervlakte (mm^2)		
58-61	mantisse	☐☐☐☐	cross-sectional area
62	exponent	☐	
63-64	13. Basisjaar	☐☐	
65-66	14. Opname-instantie (code 6)	☐☐	
67-68	15. Opname-persoon	☐☐	

APPENDIX C. continued

Vertrouwelijk. Alleen voor personeel aangewezen volgens EPL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Emissiebestand lucht *Emission file (air)*

1- 5	1. Recordcode	3 9 0 9 6	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	*plant number*
13-15	4. Bronnummer	☐☐☐	*source number*
16-19	5. Volgnummer (niet invullen)	☐☐☐☐	
20-22	6. Installatienummer	☐☐☐	*installation number*
23-24	7. Apparaatnummer	☐☐	*apparatus number*
25-28	8. Stofnummer	☐☐ ☐☐	*code of substance emitted*
29	9. Emissievorm (code 14)	☐	*code of shape of emission (gas, droplets, dust (fine) etc.)*
	10. Massastroom (mg/h)		
30-33	mantisse	☐☐☐☐	*amount of emission*
34	exponent	☐	
35-36	11. Emissiemodus (code 15)	☐☐	*code of activity (e.g. combustion of production cleaning)*
37-38	12. Capaciteit (10%)	☐☐	*production level*
	13. Volumestroom (cm^3/h)		
39-42	mantisse	☐☐☐☐	*amount of gas containing emissio*
43	exponent	☐	
44-46	14. Temperatuur (°C)	☐☐☐	*temperature*
47-48	15. Frequentie: waarde *frequency*	a ☐	*numerical*
49	eenheid (code 16)	p	*code*
50-51	16. Tijdsduur : waarde *duration*	b ☐	*numerical*
52	eenheid (code 17)	g	*code*
53-56	17. Tijdsopgave: maand *month*	☐ c t/m ☐ d	
57-58	dag *day*	e t/m f	
59-62	uur *hour*	☐ g tot ☐ h	

temporal resolution :

if a = 1
p = day
b = 8
g = hour
c = 01
d = 12
e = 1
f = 5
g = 8
h = 17

then emission
1 x per day
during 8 hours
from Jan. - Dec. (i.
from Mon. - Fri. (in
from 8.00 - 17.00 h

APPENDIX C. continued

	18. Gasreiniging: pollution control				
63-70	type (code 18) equipment code	☐☐	☐☐	☐☐	☐☐
71-82	rendement (0,1%) efficiency	☐☐,☐	☐☐,☐	☐☐,☐	☐☐,☐
83-86	bestemming (code 25) destination of obtained material	☐	☐	☐	☐
87-88	19. Oorzaak wijziging (code 19)	☐☐			
89-90	20. Soort bepaling (code 20)	☐☐	code for registration technique:		
91-92	21. Basisjaar	☐☐			
93-94	22. Opname-instantie (code 6)	☐☐			
95-96	23. Opname-persoon	☐☐			

code for registration technique:
{use of emission factor
{source test
{etc.
who? : {TNO
{plant officials
{other officials

Vertrouwelijk. Alleen voor personeel aangewezen volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Bronnenbestand water

Source file (water)

1- 5	1. Recordcode	3 9 0 9 7	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	plant number
13-15	4. Bronnummer	☐☐☐	Source number
16	5. Bronsoort (code 21)	☐	Source category code
17-36	6. Naam	☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐	
37-39	7. Ligging: hor. km	☐☐☐	
40-41	10 m	☐☐	location
42-44	ver. km	☐☐☐	
45-46	10 m	☐☐	
47-49	8. Gemeente (code 2)	☐☐☐	municipality code
50-58	9. Bestemming	☐☐☐☐☐☐ ☐☐☐	destination code [1]
59-62	10. Ont.oppervl.water (code 26)	☐☐☐☐	code for receiving surface water
63-64	11. Waterkwaliteitsbeheerder (code 22)	☐☐	has to do with water quality legislation
	12. Volumestroom (m^3/jaar)		
65-68	mantisse	☐☐☐☐	amount of wastewater
69	exponent	☐	
70-71	13. Basisjaar	☐☐	
72-73	14. Opname-instantie (code 6)	☐☐	
74-75	15. Opname-persoon	☐☐	

[1] code (not category code) for:
- hydrological defined area
- soil
- waste-water treatment plant
- other installations
- sewer

APPENDIX C. continued

Vertrouwelijk. Alleen voor personeel aangewezen
volgens ERL-regels. Geen basis voor heffingen.

EMISSIEREGISTRATIE

Emissiebestand water *Emission file (water)*

1- 5	1. Recordcode	3 9 0 9 9	
6	2. Mutatiesoort (code 1)	☐	
7-12	3. Bedrijfsnummer	☐☐☐☐☐☐	*plant number*
13-15	4. Bronnummer	☐☐☐	*Source number*
16-19	5. Volgnummer (niet invullen)	☐☐☐☐	
20-22	6. Installatienummer	☐☐☐	*installation number*
23-24	7. Apparaatnummer	☐☐	*apparatus number*
25-28	8. Stofnummer	☐☐ ☐☐	*Code of substance emitted*
29	9. Emissievorm (code 23)	☐	*Code of shape of emission (Solution, Suspension etc.)*
	10. Massa- of warmtestroom (mg/h of Watt)		
30-33	mantisse	☐☐☐☐	*amount of emission*
34	exponent	☐	
35-37	11. Herleiding tot peiljaar(%)	☐☐☐	
38-39	12. Emissiemodus (code 15)	☐☐	*Code of activity (e.g. production, cleaning ;*
40-41	13. Capaciteit (10%)	☐☐	*production level*
	14. Volumestroom (l/h)		
42-45	mantisse	☐☐☐☐	*amount of wastewater containing emission*
46	exponent	☐	
47-48	15. Frequentie : waarde *frequency*	☐☐ *numerical*	*temporal resolution*
49	eenheid (code 16)	☐ *code*	
50-51	16. Tijdsduur : waarde *duration*	☐☐ *numerical*	
52	eenheid (code 17)	☐ *code*	
53-56	17. Tijdsopgave: maand *month*	☐☐ t/m ☐☐	
57-58	dag *day*	☐ t/m ☐	
59-62	uur *hour*	☐☐ tot ☐☐	
	18. Waterzuivering: *pollution control*		
63-70	type (code 24) *equipment code*	☐☐ ☐☐ ☐☐ ☐☐	
71-73	rendement (0,1%) *efficiency*	☐☐ ☐	
74-75	19. Oorzaak wijziging (code 19)	☐☐	
76-77	20. Soort bepaling (code 20)	☐☐	*Code for registration technique :*
78-79	21. Basisjaar	☐☐	*{use of emission factor source test etc.*
80-81	22. Opname-instantie (code 6)	☐☐	
82-83	23. Opname-persoon	☐☐	*who? : {TNO plant official other officials*

U.S. EPA REGIONAL OXIDANT MODEL FOR THE TRANSPORT OF PHOTOCHEMICAL OXIDANTS AND THEIR PRECURSORS*

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STRUCTURE OF THE MODEL AND ITS INPUT DATA PROCESSOR NETWORK

The U. S. Environmental Protection Agency (EPA) Regional Oxidant Model (ROM) is designed to simulate hourly averaged concentrations of photochemical pollutants over periods of several days on a three-dimensional spatial grid that is 10^3 km in size, with a horizontal resolution of about 18 km x 18 km. This model is intended to assist the states in formulating emission control plans that will bring air quality into compliance with Federal standards. To provide reliable service in this capacity, the model was structured to consider all of the chemical and physical processes that are known, or presently thought, to affect air pollutant concentrations over several-day/1,000-km-scale domains. Among the processes included are:

- Horizontal transport;
- Photochemistry, including the very slow reactions;

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*This paper has been reviewed by the Environmental Sciences Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

†On assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce.

- Nighttime chemistry of the products and precursors of photochemical reactions;
- Nighttime wind shear, stability stratification, and turbulence "episodes" associated with the nocturnal jet;
- Cumulus cloud effects that vent pollutants from the mixed layer, perturb photochemical reaction rates in their shadows, provide sites for liquid-phase reactions, influence changes in the mixed-layer depth, and perturbhorizontal flow;
- Mesoscale vertical motion induced by terrain and horizontal divergence of the large-scale flow;
- Terrain effects on horizontal flows, removal, diffusion;
- Subgrid-scale chemistry processes resulting from emissions from sources smaller than the model's grid can resolve;
- Natural sources of HCs, NO_x, and stratospheric O_3; and
- Wet and dry removal processes (e.g., washout and deposition).

Based on analyses of aircraft O_3 and NO_x measurements and of meteorological variables over the Northeastern United States, it was concluded that incorporating all of the processes listed above would require, at the very least, a three-level model. One level would be assigned to the surface layer, a second level to the remainder of the daytime mixed layer, and a third to the layer atop the mixed layer where convective clouds are often present. However, in order for only three levels to suffice, each would have to be able to expand and contract locally in response to changes in the meteorological phenomena that each layer was intended to simulate.

The EPA regional model is designed in such a way. In addition to three layers of variable thickness, the model possesses a shallow surface layer, called Layer 0, which is adjacent to the ground. This layer handles surface deposition and subgrid-scale chemistry phenomena, and the concentration values computed in this layer represent ground-level conditions. The model is shown in Figures 1 and 2. A more detailed specification is provided in the appendix and a complete description is available in Lamb (In press, 1982).

A model as comprehensive as this one is necessarily complicated and a rather sizeable team of people was needed to develop, test, and operate it. Moreover, numerous mathematical descriptions have been proposed for the physical and chemical processes cited earlier that affect air pollutant concentrations, and new and improved descriptions are continually being developed. In view of all these considerations, the EPA model was structured in a highly modular form to aid the division of labor in the model development and maintenance, to simplify the task of trouble-shooting, and to facilitate the interchange of existing and future methods of describing the various physical and chemical processes that the model represents.

The overall structure of the model is illustrated in Figure 3. The box labeled CORE represents the computer analog of the differential equations describing the governing processes. This analog is in a very primitive mathematical form in the sense that its inputs are matrices and vectors whose elements are composites of meteorological parameters, chemical rate constants, etc. For example, the link between CORE and the output of the module labeled CHEM, which contains the analog of the chemical kinetics scheme, consists of two vectors, P and Q, each of length N, where N is the total number of chemical species simulated. The n-th element of P is the net production rate of species n due to its chemical interaction with all other species, and the n-th element of Q is the net rate of destruction of species n due to its chemical

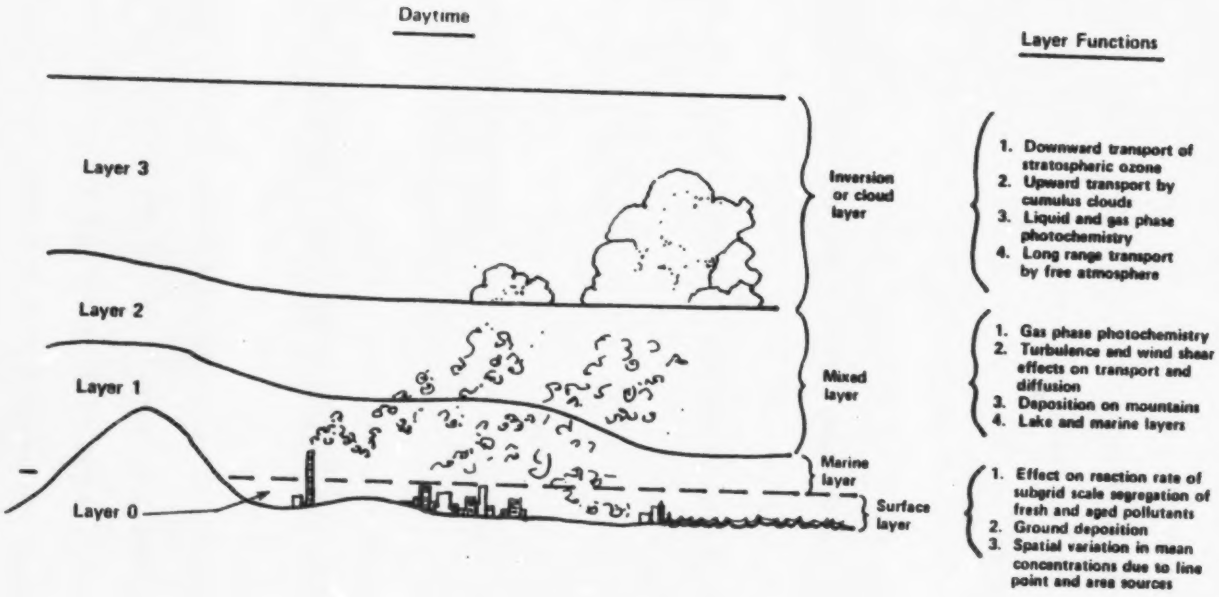
Daytime
Layer Functions
Layer 3
Layer 2
Layer 1
Layer 0
Inversion or cloud layer
Mixed layer
Marine layer
Surface layer
1. Downward transport of stratospheric ozone
2. Upward transport by cumulus clouds
3. Liquid and gas phase photochemistry
4. Long range transport by free atmosphere
1. Gas phase photochemistry
2. Turbulence and wind shear effects on transport and diffusion
3. Deposition on mountains
4. Lake and marine layers
1. Effect on reaction rate of subgrid scale segregation of fresh and aged pollutants
2. Ground deposition
3. Spatial variation in mean concentrations due to line point and area sources

Figure 1. The "dynamic" layer structure of the ROM and the daytime phenomena each layer is designed to treat.

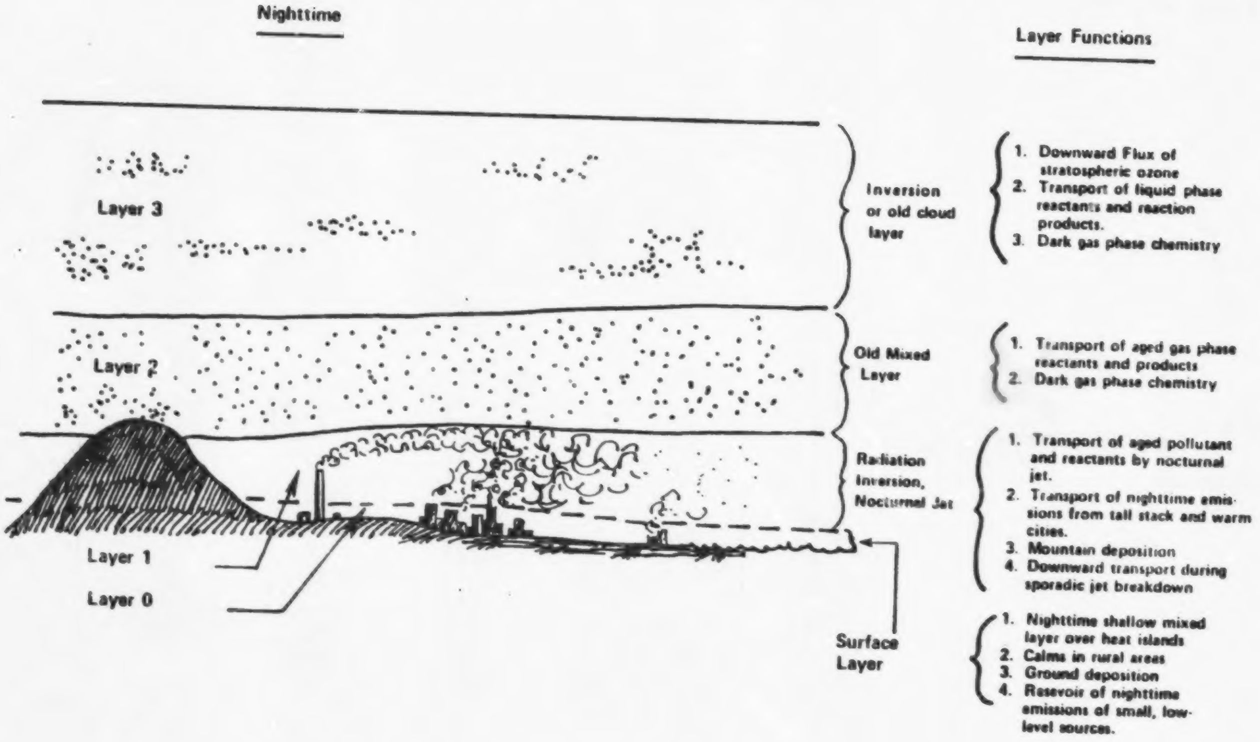
Nighttime
Layer Functions
Layer 3
Layer 2
Layer 1
Layer 0
Inversion or old cloud layer
Old Mixed Layer
Radiation Inversion, Nocturnal Jet
Surface Layer
1. Downward Flux of stratospheric ozone
2. Transport of liquid phase reactants and reaction products.
3. Dark gas phase chemistry
1. Transport of aged gas phase reactants and products
2. Dark gas phase chemistry
1. Transport of aged pollutant and reactants by nocturnal jet.
2. Transport of nighttime emissions from tall stack and warm cities.
3. Mountain deposition
4. Downward transport during sporadic jet breakdown
1. Nighttime shallow mixed layer over heat islands
2. Calms in rural areas
3. Ground deposition
4. Resevoir of nighttime emissions of small, low-level sources.

Figure 2. The "dynamic" layer structure of the ROM and the nighttime phenomena each layer is designed to treat.

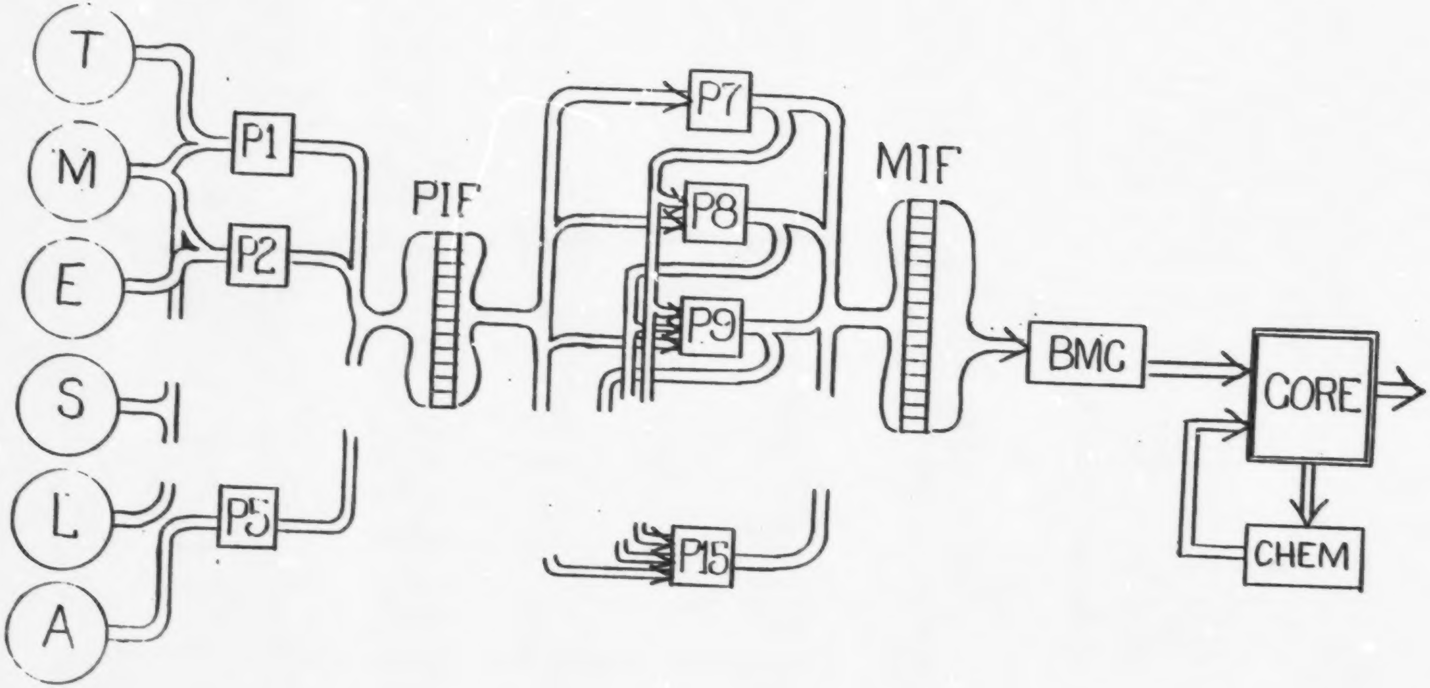
T
M
E
S
L
A
P1
P2
P5
PIF
P7
P8
P9
P15
MIF
BMC
CORE
CHEM

Figure 3. Illustration of the model and its input data processor network. Circles represent six groups of "raw" data: Topography (T), meteorological conditions (M), emissions (E), satellite cloud observations (S), land use (L), and air quality (A). These are sources of information utilized by the network of processor models, P1, P2, etc., that provide the data required by the model (CORE).

interaction with all other species. Thus, any chemical kinetics mechanism can be incorporated into the model as long as it is expressed in a form that is compatible with the vector interfaces that link CORE with the chemistry module CHEM. The rate equations that describe the chemical reactions are handled in their differential form in CORE; pseudo-steady-state approximations are not used. In the current phase of the model development, the kinetics mechanism developed by Demerjian and Schere (1979) is used, which includes some 35 reactions for 23 chemical species.

The remainder of the inputs required by CORE are prepared by the module designated BMC (b-matrix compiler) in Figure 3, which performs essentially the same task that language compilers perform in computers. The BMC translates the parameters in the model input field (MIF) into the matrix and vector elements that are required to operate the algorithms in CORE. These parameters consist of the layer thicknesses, horizontal winds in each layer, interfacial volume fluxes, deposition velocities, etc.

The variables in the MIF are supplied in turn by a network of interconnected processors (labeled P7, P8, etc. in Figure 3), several of which are rather complex models in themselves. These processors generate the wind fields, the interfacial surfaces that separate the layers, turbulence parameters, and source emissions. Their inputs consists of information generated by other processes in the network and of partially processed raw data that are transferred through the processor input file (PIF). The specific data requirements of the processor network are described later.

Each of the processors, including CHEM, can be replaced by any module that performs the same functions and that is compatible with the data channel interfaces, whose specifications are a fixed part of the network. Therefore, no specific method of treating meteorological variables is an integral part of the model. Only the data channels, the BMC, and the mathematical equations in CORE that describe the volume-averaged concentrations of pollutants within each of the model's four layers are firm parts of the system.

REQUIRED MODEL RESOLUTION AND CURRENT DATA

The ROM typically requires data in gridded form for the entire modeling region, which currently extends from 69° to 84° west longitude and from 38° to 45° north latitude. The grid system is defined in curvilinear coordinates, with a grid spacing of 1/4° east/west longitude and 1/6° north/south latitude, resulting in 2,520 grid cells (60 columns and 42 rows) of slightly varying area, approximately 18.5 km x 18.5 km. The model generally requires hourly temporal resolution for most input parameters, even though the standard model time step is 30 min. In the absence of more temporally resolved raw data, the model preprocessors use a variety of interpolation schemes to provide the final required temporal resolution.

Different types, forms, and resolutions of raw data must be standardized and combined into the consistent, chronologically ordered data sets required by the preprocessors. Each raw data set is transformed into a standard format associated with that data type and all data of similar type are merged and sorted. Thus, consistent quality control checks, graphical analysis, and standardized input/output procedures can be applied.

Much of the raw data were obtained from various national data bases. However, the complexity and scope of the ROM required more detailed and extensive information than those standard data bases provided. Therefore, EPA conducted several special field programs to gather the ambient air quality and meteorological data required for ROM development, evaluation, and application, including: the Northeast Corridor Regional Modeling Project (NECRMP), the Northeast Regional Oxidant Study I and II (NEROS), and the Persistent Elevated Pollution Episode (PEPE) Study. (For details, see Freas 1983; Possiel and Freas, 1983; and Possiel et al., 1982). In further discussions, NECRMP will be used to designate data collected during any of these field studies.

Upper Air Data

The two major U.S. sources of upper air data for ROM were the National Weather Service (NWS) radiosonde data obtained from the National Center for Atmospheric Research and the NECRMP radiosonde, pibal, and acoustic sounder data. Upper air data for 24 Canadian stations were obtained from the National Meteorological Center (NMC). Ten NWS stations fell within the model domain, and 14 additional sites surrounding the region and extending into Canada were required to resolve boundary conditions. The NWS stations are evenly distributed across and around the modeling region. The NECRMP upper air network consists of six radiosonde sites, six pibal sites, and four sodar locations aligned along the urban corridor from Virginia to Massachusetts. Typical measurements include vertical profiles of pressure, temperature, dew point temperature, dew point depression, relative humidity, wind speed, and wind direction at all mandatory and significant levels (pressure altitudes), up to 100 mbar for NWS soundings and 700 mbar for NECRMP releases. The NWS normally releases radiosondes at 12-h intervals; however, several selected sites increased their release frequency to 6-h intervals during the special field study periods--August 1979 and June, July, and August 1980. NECRMP releases occurred in the early morning, mid-morning, and early afternoon. Vertical profile data were also available from aircraft flights, tetroons, small and large tethered balloons, 3-D sodar, and minisondes.

In addition to hourly gridded values in the three layers for all listed parameters, the preprocessors required vertical wind profiles (50-m resolution) for each radiosonde release to calculate flow fields for layer-averaged winds. Station elevations were also necessary.

Surface Meteorology

Hourly surface meteorological data within the modeling region were compiled from three sources: (1) approximately 160 NWS and NMC-supplied Canadian sites, (2) 41 NECRMP sites, and (3) 27 SAROAD (Storage and Retrieval of Aerometric Data) sites. The NWS and NMC data obtained from the NOAA Techniques Development Laboratory encompass all of North America and include hourly values for temperature, dew point temperature, wind direction, wind speed, pressure, sky cover, ceiling, cloud amounts, and cloud heights. The NWS sites are evenly distributed across the region. The NECRMP and SAROAD sites report hourly wind speed, wind direction, ambient temperature, and solar radiation. NECRMP sites are located along the urban northeast corridor and SAROAD sites are distributed somewhat more evenly. Surface meteorological data were available for August 1979 and July-August 1980. Station elevations and hourly gridded values for all measurements mentioned above except solar radiation were required by the preprocessors to calculate parameters such as friction velocity, Obukov length, and heat flux. Surface parameters were gridded by using a $1/R^2$ weighting function.

Emissions Data

The EPA National Emissions Data System (NEDS) data files were not current or detailed enough for direct use in the ROM. Therefore EPA, in conjunction with States in the modeling domain, compiled an improved 1979/1980 NECRMP emissions inventory (EPA, 1982), specifically addressing the ROM requirements. Canadian emissions inventories with data ranging from 1976 to 1980 were obtained from Environment Canada to provide emission information for those portions of Canada included in the modeling domain.

Point sources are those stationary sources typically emitting greater than 100 tons of any pollutant per year. Annual U.S. emissions for point sources are reported for NO_x, VOCs, CO, SO_x, and TSP. Primary emphasis in terms of data collection and quality assurance is placed on VOCs and NO_x. Source locations are resolved to the nearest 100 m, and information on individual stack diameter, temperature, exhaust flow rate, and height are available. Emissions are reported for approximately 1,400 source classification codes (SCC). For electric utilities, fuel- and State-specific seasonal factors are calculated from power generation statistics, and hourly factors are derived from hourly power plant fuel use data. Other point source categories rely on plant-specific operating data for temporal resolution of emissions. Uniform distributions are assumed in the case of missing operating data (EPA, 1983).

The Canadian inventory contains the same types of pollutant emission and stack parameter information. The annual data on Canadian point source emissions, however, are reported for 62 different standard industrial classification (SIC) codes, and seasonal information is available for NO_x and SO_x only. Therefore, U.S. temporal allocation factors will be used to distribute most point emissions until further temporal information can be obtained.

U.S. area sources are typically mobile sources and small stationary sources individually emitting less than 100 tons of pollutant per year. Annual county-level VOC and NO_x emissions data are available for 54 area source categories. Primarily based on information gathered from the literature and previous studies, seasonal, daily, and hourly allocation factors were developed for all area source categories. County emissions were apportioned to the model grid system according to the known distribution of surrogate indicators such as housing, population, urban land, agricultural land, composite forest, land area, airport, and park locations.

Annual Canadian area source data were reported for all five pollutants (NO_x, VOCs, CO, SO_x, TSP), on the Canadian polar stereographic grid system used by the Canadian Meteorological Center (CMC). The side length of a grid is 127 km. Currently, only population data have been obtained for finer spatial resolution. However, additional surrogate information will be available from Canada in the near future. Only total emissions per pollutant are reported for each CMC grid, but percentage contributions from the 54 area source categories have been calculated.

The current chemical mechanism in the ROM expects VOC emissions to be disaggregated into four reactive classes: olefin, paraffin, aldehyde, and aromatic. The speciation methodology, which makes use of species profiles associated with process-related groups of point and area source categories, is flexible enough to generate factors to speciate reported VOC emissions into any chemical classification scheme required for the regional model. NO_x is also split into its NO and NO_2 components, based on species profile information. Hourly gridded values for these reactive HC classes, NO_x related species, CO, and initial estimates of the remaining species treated in the current chemical mechanism are required for model operation.

Land Use Data

A National Land Use and Land Cover Inventory (Page, 1980) was compiled by the EPA Environmental Monitoring Systems Laboratory for the specific latitude/longitude based grid system used in ROM. The land use and land cover data were derived from U.S. Geological Survey maps and Landsat imagery acquired during the periods July 23 through October 31, 1972, and January 1 through March 15, 1973.

The percentage of total land use and land cover in each grid cell is available for 10 categories:

- urban land
- agricultural land
- range land
- deciduous forest land
- coniferous forest land
- mixed forest land (including forested wetland)
- water
- land falling outside the study area
- nonforested wetland
- mixed agricultural land and range land

The inventory extends from 105° to 65° west longitude and 20° to 50° north latitude. The land use and land cover percentages are required by a ROM preprocessor to calculate deposition resistances and surface roughness. These data are also used in the spatial allocations of emissions inventories.

Topography Data

U.S. Air Force average elevation data were obtained from the National Center for Atmospheric Research (NCAR). Data consisted of mean elevation for global areas of 1° latitude by 1° longitude in 30-min components, with 5-min by 5-min areas for Europe, a portion of North Africa, and North America, excluding Alaska and parts of the Northwest Territories. Raw data for 5-min by 5-min areas are averaged for one model grid cell (15 ft x 10 ft) and then smoothed over a nine-cell grid area. The smoothed elevation data and the local maxima are used to incorporate terrain effects on surface deposition, horizontal winds, and mean vertical motion.

Cloud Cover and Radiation Data

Cloud cover data were available from two sources: the NWS surface stations and the Geostationary Operational Environmental Satellite (GOES) imagery. The NWS cloud data were described in the Surface Meteorology section above. Satellite images for 3 days in August of 1979 and 20 days during the period July 25 through August 25, 1980, were processed to obtain the following data for each model grid cell: (1) fractional coverage of all clouds, other than cumulus; (2) fractional coverage of only cumulus clouds and unobscured by higher clouds; and (3) average height of the cumulus cloud tops. Data images typically cover most of the model domain and are available four to five times per day.

Data were collected during August 1979 and July to August 1980. Total and ultraviolet radiation measurements were recorded at 10 surface monitoring sites and on approximately 15 aircraft flights. Spatial distribution is limited to the urban corridors and actual flight paths. Temporal resolution can vary from 10 min to 1 h.

Both cloud cover and radiation data can be used to vary photolytic rate constants in each grid for each model time step (30 min). The current methodology is to parameterize the effects of clouds on the solar spectrum rather than to require radiation measurements. Primarily, the hourly surface cloud data are gridded by using a 1/R interpolation with the scan radius technique. Cubic spline interpolation is used to derive the 30-min cloud cover values for calculating the gridded cloud transmissivity required to vary the photolytic rate constants.

Aircraft Data

During the 1979 field study, three aircraft were instrumented to provide continuous measurements of O_3, NO_x, SO_2, light-scattering coefficient (b-scat), temperature, relative humidity, and periodic canister samples for HC analysis. Sampling frequency ranged from 10 samples per second to one sample every 10 min, and data were collected for approximately 140 flights. The flight patterns were designed to provide horizontal and vertical distribution of measured parameters within specific air parcels as they were transported across the region.

Approximately 19 aircraft were involved in the 1980 field studies, collectively recording about 200 flight days of data during the 30-day study period. Additional measurements include dew point temperature, total and ultraviolet radiation, turbulence data, condensation nuclei, winds, sulfates, nitrates, and cloud chemistry in gas-phase, aerosol, and rain/cloud water forms. The 1980 aircraft sampling patterns were designed for several purposes: characterization of urban plumes, examination of urban plume interaction, and characterization of the regional air mass.

Ambient concentration data are required for testing the performance of the ROM and for determining boundary conditions. Other specialized data sets are being analyzed to provide improved parameterization of meteorological and chemical mechanisms currently in the ROM.

Surface Air Quality

The two major sources of surface air quality data were 79 NECRMP sites and 81 SAROAD sites. Hourly measurements of O_3, NO, NO_2, NO_x, nonmethane organic compounds, methane, and CO were recorded during July through mid-September 1980. SAROAD sites are scattered across the entire region, whereas the NECRMP sites are aligned along the urban corridor.

Additional field study measurements, primarily of NO, NO_x, and O_3, were taken at four stationary ground platforms and two mobile laboratories. Surface air quality measurements are used in conjunction with aircraft data for model boundary conditions, initial conditions, and evaluation.

MODEL APPLICATION IN DIFFERENT REGIONS

The design and implementation of the ROM theoretically provide maximum ease and flexibility for updating the model and preprocessor, such as shifting the model domain. Software modifications would be minimal if the same grid pattern (60 x 42) or a smaller grid pattern were defined over another region of interest. If, however, a larger number of rows and/or columns were necessary to define the modeling region, then approximately three person-months would be required to adequately modify and test all software components, i.e., approximately 40 independent programs.

The major effort required in either case is the acquisition and preparation of raw data applicable to the chosen region. Available data of each type discussed in the previous section must be reviewed for spatial and temporal adequacy. If required data are readily available, approximately three to four person-months of programming and meteorological skills are necessary to verify raw data and interface with existing preprocessors. If certain data sets are inadequate or nonexistent, then the additional costs of procuring supplementary data must be evaluated. The cost of independently obtaining data typically measured through national networks or special field studies (i.e., surface and upper air meteorology and air quality, and emissions) is generally very high. Individual estimates are highly dependent on actual requirements. Preparation of a land use and land cover inventory similar in scope and resolution to the present U.S. inventory would cost approximately $15,000. Topography data are currently available for all of Europe. Even if adequate annual point and area source emissions inventories were available for the specified region, the cost of developing specific temporal, spatial, and VOC speciation factors for the region must be added. The cost to EPA for the current set of factors was about $50,000. The estimated computer cost (with EPA's National Computer Center UNIVAC 1100/82) of generating a temporally and spatially resolved emissions inventory compatible with the ROM for one emission scenario is $15,000 (about $200 per hour). A single execution of the preprocessor system on the UNIVAC 1100/82 requires approximately 3 to 4 CPU hours, and a single 24-h model simulation requires approximately 10 CPU hours. Thorough testing of a modified model and preprocessor system would require a 3- to 4-mo commitment of a person highly knowledgeable in ROM theory and operation.

In summary, the application of the U.S. EPA ROM for a domain other than the current Northeastern United States is, in theory, fairly straightforward.

However, because of extensive data requirements and the complexity of the software system, the actual accomplishment of this objective requires significant resources, both in dollars and skilled personnel.

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DISCUSSION

H. van Dop: ...those simulations were without further confusion?

R. Lamb: In these, there is a weak leakage, 1 ml/s. We thought 1 ml/s would be small enough that there would be no appreciable leakage. However, after 50 h a lot of stuff can leak out, even at that slow a rate. We account for that leakage in a so-called true solution.

E. Runca: Is this a simulation then? Is there some accounting for the subgrid-scale effects?

R. Lamb: The subgrid scale effects are handling this Layer 0, the bottom layer. It is parameterized there. Taking into account the fact that some cells the sources are mines or they are some network of absorption.

One of the recent pieces of data that goes into the model is a description of how much the sources are segregated within any cell. If they are all within one point, then we can take that into account, but it is a parameterization of a subgrid effect.

E. Runca: So the parameterization takes place in the first layer?

R. Lamb: Layer 0.

E. Runca: Layer 0.

R. Lamb: Where the area emissions are in Layer 0, and the parameterization is there. So, in effect, the top of Layer 0, the stuff that goes in Layer 1 now is a function of the parameterization.

APPENDIX. QUESTIONNAIRE ON THE CHARACTERISTICS OF EXISTING REGIONAL-SCALE O_3 MODELS

Depending on whether your model is a Lagrangian or Eulerian type, answer the questions in either Part I or Part II of this questionnaire. Then, answer the questions in Part III.

I. Lagrangian Models

1. How are the horizontal wind fields derived (e.g., from a model, from r^{-n} interpolation of data, etc.)?
2. How is the trajectory of the coordinate origin computed (e.g., from wind at a fixed level)?
3. How are the effects of wind shear parameterized?
4. How is lateral diffusion parameterized?
5. How is vertical diffusion treated?
6. How are species concentrations outside the plume determined?
7. Are plumes from neighboring sources allowed to mix and react?
8. What is the vertical resolution of the model (e.g., number of levels and Δz)?
9. What numerical method is used to solve the equations that govern vertical mixing and reaction processes?
10. Does the mixed layer depth vary with travel time? If so, how is it determined?
11. Is mean (nonzero) vertical motion permitted? If so, how is the vertical speed determined (e.g., from divergence of observed horizontal winds)?
12. Is horizontal concentration variation resolved within the puff or plume?

    (a) With what resolution (give Δx and Δy)?

    (b) How many grid points in the horizontal plane?

(c) What numerical scheme is used to solve the equations governing horizontal mixing and reaction?

(d) Is horizontal variation of source emission rates resolved within the puff or plume?

(e) If horizontal concentration variations within the puff or plume are not resolved, how are the effects of these variations on reactions rates parameterized?

13. Are convective cloud effects treated?

14. Is surface deposition treated? If yes:

(a) Are spatial variations in the rate permitted?

(b) Are temporal variations permitted?

15. Are terrain effects treated? Are land use effects treated?

16. Over how large an area are trajectories calculated?

17. What is the temporal resolution of the model?

18. How much machine time is required to compute 24-h averaged O_3 measurements at a single receptor? (Which computer?)

19. Has the model been tested? If yes, cite report.

20. Is the model available for use? If not, when will it be available?

II. Eulerian Models

1. What is the temporal resolution (i.e., Δt)?

2. What is the horizontal resolution (Δx and Δy)?

3. What are the present horizontal dimensions of the model domain?

4. What is the vertical resolution (i.e., number of levels and Δz)?

5. What is the elevation of the top level of the model?

6. How are the horizontal wind fields derived (e.g., from a model, from r[illegible] interpolation of data, etc.)?

7. What numerical scheme is used to treat the horizontal transport terms of the governing equations? If pseudo-spectral, how are inflow boundary conditions handled?

8. Is mean (nonzero) vertical motion simulated? If so, how is the mean vertical speed determined (e.g., from divergence of horizontal winds)?

9. What numerical method is used to treat the vertical transport and diffusion terms in the governing equations?

10. How is horizontal diffusion parameterized?

11. How is vertical diffusion treated?

12. Is the mixed layer depth variable in space?

    (a) Is it variable in time?

    (b) How is the depth determined?

    (c) How is it simulated in the model (e.g., in the form of the $K_z(z)$ profile)?

13. Are convective cloud effects treated?

14. Are terrain effects included?

15. Is surface deposition parameterized?

    (a) Are the deposition velocities variable in space?

    (b) Are they variable in time?

16. Are rainout and washout processes included?

    (a) Are the rates spatially variable functions?

17. How much machine time is required to compute 24-h averaged O_3 measurements? (Express estimate in terms of machine seconds divided by the total number of surface grid points in the model domain, i.e., s/grid point.

18. How much computer memory would be required for a simulation of a 10^3 x 10^3 km region with a mesh size of 20 km? How many species does this estimate include? Which computer?

19. Has the model been tested? If yes, cite report.

20. Is the model available for use? If not, when will it be available?

III. Chemistry

1. Is the chemical kinetics scheme a fixed part of the model or can it be interchanged with other schemes?

   If fixed:

   (a) List the species that are treated as dependent variables.

   (b) List any species whose concentrations are prescribed.

   If interchangeable:

   (c) Which schemes have been tested?

   (d) List the species that are treated as dependent variables in the scheme currently used.

   (e) List any species whose concentrations are prescribed.

   (f) What is the largest number of species that the model can accommodate?

2. Is the pseudo-steady-state approximation used in solving the chemical rate equations?

3. Are the effects of subgrid-scale concentration variations parameterized?

4. How are emissions of major point sources treated?

5. Does the model provide any measure of how much the concentration at a point may differ from the cell averaged value?

6. Do the photolytic rate constants vary in space as a function of cloud cover?

Responses to Part II of Questionnaire on the ROM

1. Temporal resolution = 30 min.

2. Horizontal resolution = 1/4° longitude x 1/6° latitude.

3. Model dimensions are 60 x 42 cells ≈ 1,100 km x 780 km.

4. Vertical resolution is by four layers whose thicknesses vary in space and time. Nominal values of the elevations of the tops of each layer under clear sky conditions would be as follows: Layer 0--30 m, Layer 1--300 m, Layer 2--1,500 m, Layer 3--2,000 m.

5. Top surface of the model is variable in space and time and is set at each grid point to be just above the top of any convective clouds present in that cell.

6. Horizontal wind fields can be derived by any method desired. Currently the horizontal flows are generated in the form of function sets (Lamb, 1982b) derived jointly from observations and physical laws.

7. Horizontal transport and diffusion terms in the governing equations are approximated by the explicit, biquintic scheme described in Chapter 9 of Lamb (1982a).

8. Mean vertical motion is included in the model. The method of estimating its magnitude at each point in space and time is optional. The calculation is presently based on the continuity equation and computed divergence in the horizontal wind.

9. Vertical transport and diffusion are simulated by an analytic solution of the linearized vertical equations (see Chapter 9 of Lamb, 1982a).

10. Horizontal diffusion due to small-scale wind fluctuations, viz. convective and mechanical boundary-layer turbulence, is approximated using K theory. The larger mesoscale fluctuation effects are represented by the function sets of velocity fields, referred to above and discussed in Chapter 7 of Lamb (1982a).

11. Vertical diffusion is treated by volume fluxes of material between layers caused by turbulent fluctuations in the vertical wind.

12. The mixed-layer depth is variable in space and time. It is represented in the model by the combined thicknesses of Layers 0, 1, and 2. When convective clouds are present, the top of Layer 2 is defined as the lifting condensation level and the effective mixed layer extends into Layer 3. The method of estimating the mixed-layer depth is optional. Currently, it is based on measured vertical profiles of temperature and dew point and on the estimated vertical profile of potential vorticity.

13. Convective clouds are treated explicitly, including vertical material transfer, attenuation of sunlight, etc.

14. Terrain effects on surface deposition, horizontal winds, and mean vertical motion are included.

15. Surface deposition velocities are computed at each cell and each hour by using the local friction velocity and surface deposition resistences, estimated from land use data and empirical studies.

16. Rainout and washout are not included at present, but incorporation of these processes would be straightforward.

17. Approximately 10 h of UNIVAC 1,182 time is needed to simulate 1/2-h O_3 at 2,520 surface grid points over a 24-h period. Concentrations of 22 other species are also provided at all points and time steps. This is approximately 15 s per grid point for a 24-h simulation.

18. Total computer memory requirement for 23 species; 7,560 grid points (3 layers at 2,520 points per layer) is 110 K words. (Note: Layer 0 is handled diagnostically and does not require memory space.)

19. Analytical tests of the model are in progress, i.e., comparisons of the predicted values with exact solutions.

20. The model is not available for use.

Responses to Part III of Questionnaire on the ROM

1. The chemical kinetics scheme is interchangeable.

   (c) The scheme of Demerjian and Schere (1979) is currently being used.

   (d) This scheme treats 23 species: NO, NO_2, O_3, paraffin, olefin, aldehyde, aromatics, CO, HNO_2, HNO_3, PAN, RNO_3, H_2O_2, O, NO_3, HO, HO_2, HO_4N, RO, RO_2, R_2O, $R1O_2$, R_2O_2.

   (e) No species values are prescribed in the rate equation.

   (f) The largest number of species that the model can handle is limited only by the machine time one is willing to buy.

   (g) Simulations of 2 days have been done. Longer simulations are anticipated.

2. (a) No pseudo-steady-state assumptions are used.

   (b) Nighttime chemistry kinetics is the same as the daytime, with the photolytic rate constants all set to zero.

   (c) Nighttime wind shear, stability stratification, and turbulence are simulated.

3. (a) Subgrid-scale chemistry phenomena are parameterized in the bottom layer only, i.e., Layer 0.

   (b) Subgrid-scale effects are those due to the segregation of fresh emissions from line sources and aged pollutants brought down to ground level by turbulence.

4. Emissions of major point sources are not now treated in a rigorous way. A scheme will be developed.

5. Layer 0 contains the means of estimating how much the concentration at any point in that cell might differ from the cell-averaged value.

6. (a) Rate constants vary in space and time due to sun angle and cloud cover variations.

   (b) Convective cloud effects on vertical material fluxes, mixed-layer depth, and photochemical reaction rates are treated. Liquid-phase chemistry is not treated at this time.

7. Natural emissions from vegetation are estimated on the basis of biomass levels in each grid cell, temperature, and time of day. Stratospheric O_3 entrainment into the model domain is parameterized by using estimates of O_3 concentration aloft and mean vertical air speed.

REGIONAL MODEL FOR OXIDANTS: THE NORWEGIAN LAGRANGIAN LONG-RANGE TRANSPORT MODEL WITH ATMOSPHERIC BOUNDARY LAYER CHEMISTRY*

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INTRODUCTION

The Norwegian Lagrangian long-range transport model for oxidants represents the joint effort of three institutions--the Institute of Geophysics at the University of Oslo, the Norwegian Meteorological Institute, and the Norwegian Institute for Air Research. The model is actually a combination of a chemical model describing the gas-phase chemistry of HCs, NO_x, and SO_2, together with a simple parameterization of the gas-phase/aerosol-phase interaction, and a meteorological model describing the long-range transport of air pollutants (Eliassen et al., 1982). The chemical model was developed at the University of Oslo (Hesstvedt et al., 1978; Hov et al., 1978a; Isaksen et al., 1978; Derwent and Hov, 1979, 1980a). This model has been used to demonstrate that O_3 can survive in the atmospheric boundary layer for a week or more and thus can be transported over long distances (Hov et al., 1978b). The meteorological model was originally developed as part of an OECD study on the long-range transport of air pollutants (OECD, 1977; Eliassen, 1978). It is now being applied at the Norwegian Meteorological Institute in connection with the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP), which is being sponsored by the United Nations Economic Commission for Europe (ECE).

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

The EMEP project is divided into two areas, chemical activities and meteorological activities. The Norwegian Institute for Air Research (NILU) serves as the coordinator for the chemical activities; two other centers, one in Moscow and the second at the Norwegian Meteorological Institute (NMI), coordinate the meteorological activities. Some of the modeling efforts conducted for EMEP have been described by Eliassen and Saltbones (1983), and reports on monitoring and interpreting the long-range transport of oxidants in Norway have also been published (Schjoldager et al., 1978). Inventories on NO_x and SO_2 emissions in Europe have also been reported by Semb (1979) and by Dovland and Saltbones (1979).

In 1978, a planning conference on the long-range transport of photochemical oxidants was held by NILU under OECD patronage. The conference organizers presented a list of questions on the characteristics of the model. The answers to these questions are discussed throughout this text where appropriate.

It is important to consider the construction of a model as a continuous development, and the questions to be answered by the model should determine its formulation. Thus, a model should be formulated with such flexibility that the most important processes involved in answering a question can be identified and quantified. Excessive complexity inevitably leads to several problems, such as a heavy investment in computer capacity and model management. Such complexity may be distracting when the impact of the various processes included in the model must be assessed. The complexity may thus disguise the poor performance of some components of the model.

MODEL DESCRIPTION

Meteorological Model

The Norwegian model presently employs 96-h, 850-mbar back trajectories, which are assumed to be trajectories of polluted boundary-layer air parcels. The pollutants are assumed to be completely mixed vertically throughout a boundary layer of variable depth; the concentrations of the various species are therefore functions of the horizontal coordinates and time. The same assumption is made for the horizontal wind. The top of this well-mixed layer is assumed to be a material surface, through which no mass transport takes place. Lateral diffusion is neglected, because the emission data are given in a 150-km grid; hence, finer details in the concentration fields are already smoothed out.

In episode studies based on short sampling periods (i.e., periods much less than 24 h), the rate at which instantaneous pollutant releases spread horizontally may be an important parameter to consider (Eliassen, 1982). For a 24-h sampling period, which is used for sulphur species in EMEP, the instantaneous diffusion of pollutant releases is dominated by the diffusion due to sampling time. The "synoptic swinging" (Smith, 1979) of trajectories is then the dominating factor in plume spread (Eliassen, 1982).

During transport, pollutants are emitted into the air parcel according to the NO_x, HC, and SO_2 emissions maps, and the chemical reactions between the various species proceed continuously. At the specified receptor points, instantaneous concentrations are predicted upon arrival of a trajectory.

A receptor-oriented trajectory model, such as the one outlined above, has an important advantage over a source-oriented trajectory model: Nonlinear air chemistry can be included fairly easily. This is not true for a source-oriented model in which the individual "puffs" emitted from the different sources are followed separately. In practice, "puffs" emitted from different sources at different times may overlap, and the chemical species present in the overlapping should then be allowed to interact in a nonlinear chemical scheme. Because of the nonlinearity, however, the distribution of pollutants back into the individual puffs after the chemical interaction has occurred is indeterminate. If one tries to handle this situation by letting all overlapping puffs merge, the result is, in fact, a receptor-oriented model.

In a receptor-oriented model, the horizontal resolution of the concentration fields is determined by the choice of emission grid and the density of trajectory arrival points. The horizontal resolution that can be achieved is, to a large extent, a function of how well the horizontal distribution of the emissions is known.

If an Eulerian approach were taken, such problems would not arise. In principle, one could construct multilayer models that would take into account such factors as the combined effect of wind shear and vertical diffusion. This situation would be difficult to handle in Lagrangian models. However, Lagrangian or trajectory models have one important advantage over Eulerian models: In Lagrangian models the integration of the equations is reduced to an ordinary time integration along selected trajectories. In terms of the numerical methods and the computer capacity required, this is a much simpler problem to handle than a complete Eulerian integration over a large geographical area.

Trajectory positions were calculated every 2 h by using the method of Petterssen (1956). The calculations were based on wind observations made at the 850-mbar level at 0000, 0600, 1200, and 1800 GMT. The observed wind data were analyzed objectively in the 150-km grid indicated in Figure 1. This is the EMEP grid, which covers a net of 37 x 39 grid squares for an area of 150 km x 150 km^2. The temporal resolution of the model is adjustable, but as mentioned above, currently instantaneous concentration fields were calculated every 6 h. In regions where wind observations were scarce, such as areas over seas, the final wind analysis was heavily influenced by the quasi-geostrophic balanced wind produced by NMI on a 300-km grid as part of its weather prediction routine.

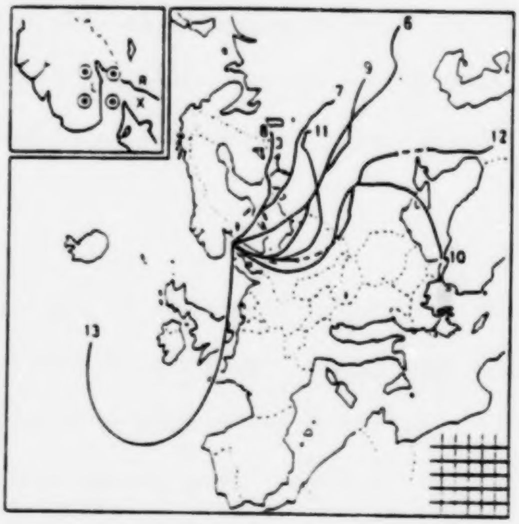


Figure 1. Air trajectories (850 mbar) followed for 4 days, arriving at one of the five receptor points at 1200 GMT, April 6-13, 1979. The arrival dates are indicated at the starting point of the trajectories. The broken line represents the high NO_x and HC emission rates for the trajectory arriving April 12. Ozone was measured at Langesund (L). The four receptor points used in the model calculations of O_3 are denoted by circles. Sulphur dioxide and particulate sulphate were measured at Rorvik (R). The size of the 150-km grid squares is shown in the lower right-hand corner. The meteorological variables are given in the same grid.

Alternative trajectories may be calculated by backing the analyzed 850-mbar winds (e.g., 15°) and by reducing the wind speed (e.g., 90%) when the radiosonde observations in the vicinity of the trajectories indicate a significant turning of the wind with height and change in wind speed. At present, the mixing of pollutants associated with the diurnal variation of the atmospheric boundary layer is not described. This would require a model with several layers, in which "new" pollutants would be emitted into a shallow but growing convective boundary layer and "old" pollutants, those emitted on previous days, would be distributed within a deeper layer. The mixing height used in the Norwegian model is assumed to represent an "envelope" height, below which both old and new pollutants are mixed. The 1200-GMT mixing height was chosen for this purpose. Over most of Europe, 1200 GMT (which corresponds to 1300 or 1400 local time) is the time by which a growing convective boundary layer has nearly reached its maximum height.

This mixing height envelope is assumed to be a material surface through which no mass transport takes place. Upward or downward movements of this surface can be caused by horizontal convergence or divergence, and the density of the air is assumed to be constant. These assumptions imply that the concentrations of pollutants already present in the mixing layer should not be diluted when the mixing height along a trajectory increases. The initial concentration inputs from primary pollutant emissions are of course inversely proportional to the mixing height.

The simplified description of vertical dispersion causes errors that are difficult to quantify. The real situation is complicated and difficult to handle in models. The pollutants are not only affected by the diurnal cycle of the mixing layer, but they are also present in plumes during the initial phase of dispersion. Thus, a nonlinear chemistry scheme using volume-averaged concentrations may incorrectly describe the chemical development, especially during the initial dispersion phase.

The basic data for the mixing-height analysis were taken from radiosonde data. On the average, about 120 radiosonde reports are available within the grid. The height up to the lowest stable layer (potential temperature increasing with height) was used as the mixing height for each radiosonde report. Stable layers with a base lower than 200 m were ignored. If no stable layer was found, the mixing height was set equal to an upper limit, which was arbitrarily selected as 2,500 m. The estimated mixing heights were thereafter objectively analyzed to produce grid values at 1200 GMT. The individual trajectories were assigned new mixing heights at 1200 GMT every day, according to their position and the relevant mixing-height field. At intermediate times, it is assumed that each trajectory conserves its mixing height.

A number of difficulties are associated with the objective analysis of mixing height. For example, the mixing height estimated from a radiosonde ascent will depend on the definition of a stable layer. Recent model tests suggest that a lapse rate of about half the dry-adiabatic value, rather than the dry adiabat itself, would give a better division between stable and unstable layers (Eliassen and Saltbones, 1983).

In an experimental version of the EMEP model, which may also be adopted in the long-range transport of oxidant modeling work, the exchange of pollution between the atmospheric boundary layer and the free troposphere was modeled by Eliassen and Saltbones (1983).

Consider a parcel of boundary-layer air that follows a calculated trajectory. The air parcel starts at t_1 = 12 GMT with a mixing height h_1 from the objective analysis. Assuming that the mixing height behaves as a material surface, the air parcel will have a new mixing height h(t) after some time t, given by:

$$h(t) = h_1 + \int_{t_1}^{t} w(t)dt$$

where w(t) is the vertical velocity at h(t), taken at the position of the air parcel. The equation for h may be used to calculate the mixing height $h(t_2)$ 1 day later; i.e., $t_2 = t_1 + 24$ h. In general, $h(t_2)$ will be different from the mixing height h_2, which is available from the analyzed mixing height field at t_2.

It is now assumed that h_2 is the correct mixing height for the air parcel at t_2 and that the difference between h_2 and $h(t_2)$ arises because the mixing height is not in reality a material surface, but that there is a certain flux of air through it. If $h(t_2) > h_2$, then some boundary-layer air has been lost to the free troposphere. If $h(t_2) < h_2$, then some air from the free troposphere has penetrated the boundary layer. Using two methods, Eliassen and Saltbones estimate the vertical velcocity at h(t). In the first, the velocity is estimated from the divergence of the horizontal wind. In the second, w is estimated as that due to frictional convergence in an Ekman boundary layer (Eliassen and Saltbones, 1983).

An objective analysis of temperature, relative humidity, and absolute humidity were carried out at 0000 and 1200 GMT in the 150-km grid. The quantities analyzed were vertical averages between the surface and the 850-mbar level. A detailed description of the methods used can be found in Eliassen et al. (1979). These analyses are also based on radiosonde data.

The temperature is used to evaluate certain temperature-dependent reaction rate coefficients, such as the thermal decomposition of PAN, which influences the O_3 concentration through the production of NO_2.

The relative humidity is used as a rough indication of cloud cover, which in turn influences the calculated photodissociation rate coefficients through an "effective" albedo. Because no reflection is assumed at the ground level, any cloud cover will reduce the photodissociation rate coefficients. The parameterization shown in Table 1 was used.

The calculation of photodissociation rate coefficients, including the influence of the cloud albedo, is described below. The relative humidity also determines when certain components are removed by wet deposition.

The absolute humidity is a measure of the concentration of water vapor molecules. This is used as input for the air chemistry model, in which H_2O in part determines the OH concentration.

The individual trajectories are assigned new temperature and absolute humidity values when analyzed fields of these quantities are available, i.e., at 0000 and 1200 GMT. At intermediate positions, the temperature is estimated by linear interpolation, whereas the absolute humidity is assumed to be conserved.

TABLE 1. PARAMETERIZATION OF CLOUD COVER ALBEDO

Relative Humidity	Cloud Cover	"Effective" Albedo
>85%	1.0	0.6
75-85%	0.5	0.3
<75%	0.0	0.0

The removal rate k_d of any component due to dry deposition is:

$$k_d = \frac{v_d}{h} ,$$

where v_d is the deposition velocity and h is the variable mixing height. The calculated O_3 concentration depends heavily on the deposition velocity assumed for O_3 (Eliassen et al., 1982). Garland and Derwent (1979) report a mean deposition velocity over grassland of 0.58 cm/s by day and 0.29 cm/s by night. In assigning values to this quantity, the following factors have been considered:

- isolation of the O_3 from the surface by nighttime inversions, and
- very little, if any, uptake by the sea.

Table 2 shows a set of values for the deposition velocities of O_3 and other compounds. These values were taken from a case study on the long-range transport of oxidants to South Norway during April 1979. In this case, an additional factor was considered when assigning values to the deposition

TABLE 2. ASSUMED DRY DEPOSITION VELOCITIES FOR A CASE STUDY OF LONG-RANGE TRANSPORT OF OXIDANTS TO SOUTH NORWAY DURING APRIL 1979[a]

Component	v_d (cm/s)	
	0.1	$\psi \geq 65°N$
	0.2	$60°N \leq \psi < 65°N$
O_3 (day, land)	0.3	$55°N < \psi < 60°N$
	0.4	$50°N \leq \psi < 55°N$
	0.5	$\psi < 50°N$
O_3 (night, land)	10% of O_3 (day, land) values	
O_3 (sea)	0.0	
NO_2	0.1	Boettger et al. (1978)
PAN	0.2	Garland and Penkett (1976)
SO_2	0.8	Garland (1977)
HNO_3	1.0	Assumed
H_2SO_4	0.1	Value appropriate for submicron particles

[a] From Eliassen et al., 1982.

More recent findings indicate that a value of 0.5 cm/s may be more appropriate for the deposition velocity of NO_2 (Grennfelt, private communication; Galbally, private communication).

Wet deposition is parameterized by using the relative humidity as it varies along each trajectory. When the relative humidity exceeds 90%, precipitation is assumed and a wet deposition rate of $1 \times 10^{-4} s^{-1}$ is applied to the H_2SO_4, HNO_3, H_2O_2, and CH_3O_2H concentrations.

Ideally, the wet deposition should be calculated by storing the amount removed from the atmosphere by each precipitation episode. In an experimental version of the EMEP model, applying scavenging ratios (W) for SO_2 and sulphate resulted in wet deposition rate coefficients of the form:

$$k_w = W \frac{P_6}{h}$$

where h is the mixing height and P_6 is the objectively analyzed 6-h precipitation intensity (Eliassen and Saltbones, 1983). The objective analysis produces a smoothing so that the time resolution is 6 h and the spatial resolution is 150 km x 150 km. Such a grid square is either completely dry during a 6-h period or completely wet, with an average precipitation intensity P_6. Eliassen and Saltbones (1983) state that, in reality, the space-time area covered by a grid square will not be completely wet. The precipitation events will generally cover parts of the area, whereas other parts will be dry. The average precipitation intensity for the wet part of the space-time grid square will be larger than P_6. In the model, the trajectories will therefore have an exaggerated probability of encountering rain, but the intensity of the rain will be correspondingly lower. This reduces the probability that an air parcel will be transported for many days without encountering precipitation at all, which will therefore result in a reduced frequency of high concentrations in remote areas. Eliassen and Saltbones (1983) report a method that obviates this problem.

Chemical Model

Two types of approaches have been used in photochemical smog modeling. One applies "lumped" kinetic mechanisms, in which the various HCs are not specifically included. Instead, the HC chemistry is described by using several different classes of structure or reactivity (Hecht and Seinfeld, 1972; Reynolds et al., 1973; Hecht et al., 1974; Falls and Seinfeld, 1978). The predictions of such models depend strongly on the reaction rates and kinetic mechanisms used for the different classes of HCs. A number of parameters cannot be measured and must therefore be specified a priori. Such simple generalized mechanisms are applied due to the lack of kinetic data and the heavy demand placed on computer resources when combined schemes of transport and chemistry are to be integrated.

In recent years several models have been developed that apply specific schemes describing the photooxidation of particular HCs (Demerjian et al., 1974; Graedel et al., 1976; Hov et al., 1978a; Derwent and Hov, 1980a). The accuracy of these schemes can be determined directly from the uncertainty of the measurable rate constants involved if the air-HC mixture has been incorporated satisfactorily. The latter condition is not a trivial one, however, because samples of polluted air are known to contain hundreds of different HCs.

A simplified approach must therefore be used in constructing a model system that can be handled. The approach we have taken is to adopt a specific scheme with a limited number of precursor HCs. This scheme reproduces reasonably well the pollutant-generating capacity of a much more detailed specific scheme developed earlier to represent average UK emissions (Derwent and Hov, 1979; 1980a).

After several model runs in which several different compositions of NMHC emissions were tested, we decided to use a mixture of five different HCs to calculate pollutant transport. The selected mixture (30% C_2H_6, 20% n-C_4H_{10}, 20% C_2H_4, 10% C_3H_6, and 20% m-xylene) represents fairly well the more detailed Derwent and Hov mixture for species like O_3 and OH. A compound like PAN, however, may deviate by more than a factor of 2, due to its dependence on the peroxyacetyl radical, which is derived only from certain HCs.

There are several reasons to use such a simplified mixture: the uncertainty of the estimated NMHC emissions, the incomplete knowledge of the kinetic mechanisms, and the heavy demand on computer capacity if a more complete mixture is used.

As a result of this simplification, the chemical scheme consists of about 100 chemical reactions (including photochemical reactions) and 40 different species. A list of reactions and rate coefficients is given in Table 3. The concentration over time for all species involved in the reactions listed is calculated, with the exception of a few organic radicals that react quickly with molecular oxygen (e.g., formyl, HCO). (The O_2 concentration is prescribed as 21% of M, by volume; the CH_4 concentration is prescribed as 1.4 ppm).

The driving force behind photochemical air pollution is the stimulation of free radical production by the photolysis of light-absorbing species. Thirteen inorganic and organic species in the model are dissociated by sunlight. The data for absorption cross sections, quantum yield, and solar fluxes are taken from Calvert and Pitts (1967) and NASA (1979). The photochemical rate coefficients at the earth's surface are calculated by the method of Isaksen

TABLE 3. CHEMICAL REACTIONS AND REACTION RATE COEFFICIENTS (cm^3/molecule · s) FOR BIMOLECULAR REACTIONS (Eliassen et al., 1982).

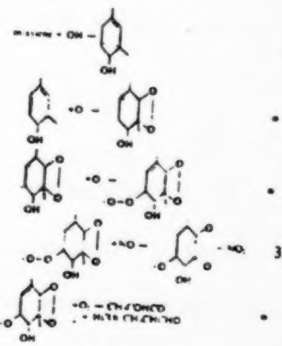
Reaction	Rate coefficient
Inorganic chemistry	
$O + O_3 + M \rightarrow O_3 + M$	$1.1 \times 10^{-34} \exp(510/T)$
$O + NO + M \rightarrow NO_2 + M$	$3.0 \times 10^{-33} \exp(940/T)$
$O^1D + M \rightarrow O + M$	3.0×10^{-11}
$H_2O + O^1D \rightarrow 2OH$	2.3×10^{-10}
$O_3 + NO \rightarrow NO_2 + O_2$	$2.3 \times 10^{-12} \exp(-1450/T)$
$O_3 + NO_2 \rightarrow NO_3 + O_2$	$1.2 \times 10^{-13} \exp(-2450/T)$
$O_3 + OH \rightarrow HO_2 + O_2$	$1.8 \times 10^{-12} \exp(-930/T)$
$O_3 + HO_2 \rightarrow OH + O_2 + O_2$	$1.4 \times 10^{-14} \exp(-580/T)$
$NO + NO_3 \rightarrow 2NO_2$	1.9×10^{-11}
$NO + HO_2 \rightarrow NO_2 + OH$	8.1×10^{-12}
$NO_2 + NO_3 \rightarrow NO + NO_2 + O_2$	$2.3 \times 10^{-13} \exp(-1000/T)$
$NO_2 + NO_3 \rightarrow N_2O_5$	$1.48 \times 10^{-13} \exp(861/T)$
$NO_2 + OH \rightarrow HNO_3$	1.1×10^{-11}
$NO_3 + H_2O_2 \rightarrow HO_2 + HNO_3$	4.1×10^{-16}
$NO_3 + NO_3 \rightarrow NO_2 + NO_2 + O_2$	$8.5 \times 10^{-13} \exp(-2450/T)$
$N_2O_5 \rightarrow NO_2 + NO_3$	$1.24 \times 10^{14} \exp(-10317/T)$
$OH + HO_2 \rightarrow H_2O$	5.1×10^{-11}
$OH + H_2O_2 \rightarrow HO_2 + H_2O$	$2.7 \times 10^{-12} \exp(-145/T)$
$OH + H_2(+O_2) \rightarrow HO_2 + H_2O$	$3.6 \times 10^{-11} \exp(-2590/T)$
$OH + HNO_3 \rightarrow NO_3 + H_2O$	8.0×10^{-14}
$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	$3.8 \times 10^{-14} \exp(1245/T)$
Sulphur chemistry	
$OH + SO_2 \rightarrow HSO_3$	1.1×10^{-12}
$CH_3O_2 + SO_2 \rightarrow SO_3 + CH_3O$	$<5 \times 10^{-17}$
$HSO_3 + O_2 \rightarrow HSO_5$	1.0×10^{-13}
$HSO_5 + NO \rightarrow HSO_4 + NO_2$	1.0×10^{-12}
$HSO_4 + O_2 \rightarrow SA^* + HO_2$	1.0×10^{-15}
$SO_3 + H_2O \rightarrow SA^*$	9.1×10^{-13}
* SA: sulphuric acid or sulphate aerosol.	
Methane chemistry	
$OH + CH_4 \rightarrow CH_3\dot{O} + H_2O$	$2.4 \times 10^{-12} \exp(-1710/T)$
$\dot{C}H_3 + O_2 \rightarrow CH_3\dot{O}_2$	5.1×10^{-13}
$CH_3\dot{O}_2 + NO \rightarrow CH_3\dot{O} + NO_2$	6.5×10^{-12}
$CH_3\dot{O}_2 + CH_3\dot{O}_2 \rightarrow CH_3\dot{O} + CH_3\dot{O} + O_2$	4.0×10^{-13}
$HO_2 + CH_3\dot{O}_2 \rightarrow CH_3O_2H + O_2$	$7.7 \times 10^{-14} \exp(1300/T)$
$CH_3\dot{O} + O_2 \rightarrow HCHO + HO_2$	1.8×10^{-15}
$OH + HCHO \rightarrow H_2O + H\dot{C}O$	$1.25 \times 10^{-11} \exp(-88/T)$
$NO_3 + HCHO \rightarrow HNO_3 + H\dot{C}O$	8.0×10^{-16}
$OH + CO \rightarrow CO_2 + H$	$1.35 \times 10^{-13} [1 + P(\text{atm})]^*$
$H\dot{C}O + O_2 \rightarrow HO_2 + CO$	5.1×10^{-12}
* P(atm): pressure in atmospheres.	
Ethane chemistry	
$C_2H_6 + OH \rightarrow \dot{C}_2H_5$	$1.86 \times 10^{-11} \exp(-1236/T)$
$\dot{C}_2H_5 + O_2 \rightarrow C_2H_5\dot{O}_2$	*
$C_2H_5\dot{O}_2 + NO \rightarrow C_2H_5\dot{O} + NO_2$	3×10^{-12}
$C_2H_5\dot{O} \rightarrow HCHO + \dot{C}H_3$	33.0
$C_2H_5O + O_2 \rightarrow CH_3CHO + HO_2$	3.7×10^{-17}
$CH_3CHO + h\nu \rightarrow CH_3 + \dot{C}HO$	3.1×10^{-5}
$CH_3CHO + OH \rightarrow CH_3\dot{C}O + H_2O$	$6.9 \times 10^{-12} \exp(258/T)$
$CH_3CO + O_2 \rightarrow CH_3CO\dot{O}_2$	*
$CH_3CO\dot{O}_2 + NO \rightarrow \dot{C}H_3 + CO_2 + NO_2$	2.6×10^{-12}
$CH_3CO\dot{O}_2 + NO_2 \rightarrow CH_3COO_2NO_2$ (PAN)	1.4×10^{-12}
$PAN \rightarrow CH_3COO_2 + NO_2$	$7.94 \times 10^{16} \exp(-12530/T)$
* Very fast reaction step.	
Ethylene chemistry	
$C_2H_4 + OH \rightarrow \dot{C}H_2CH_2OH$	$2.2 \times 10^{-11} \exp(385/T)$
$\dot{C}H_2CH_2OH + O_2 \rightarrow CH_2O_2CH_2OH$	*
$CH_2O_2CH_2OH + NO \rightarrow CH_2\dot{O}CH_2OH + NO_2$	3.1×10^{-12}
$CH_2OCH_2OH + O_2 \rightarrow HCHO + HCHO + HO_2$	*
$C_2H_4 + O_3 \rightarrow HCHO + \dot{C}H_2O_2$	$9.0 \times 10^{-15} \exp(-2560/T)$
$\dot{C}H_2O_2 + O_2 \rightarrow HO_2 + HO_2 + CO_2$	*
* Very fast reaction step.	
Propylene chemistry	
$C_3H_6 + OH \rightarrow CH_3\dot{C}HCH_2OH$	$4.1 \times 10^{-12} \exp(545/T)$
$CH_3\dot{C}HCH_2OH + O_2 \rightarrow CH_3CH\dot{O}_2CH_2OH$	*
$CH_3CH\dot{O}_2CH_2OH + NO \rightarrow CH_3CH\dot{O}CH_2OH + NO_2$	3.1×10^{-12}
$CH_3CHOCH_2OH + O_2 \rightarrow CH_3CHO + HCHO + HO_2$	*
$C_3H_6 + O_3 \rightarrow HCHO + CH_3\dot{O}_2 + HO_2 + CO_2$	$3.05 \times 10^{-15} \exp(-1900/T)$
$\rightarrow CH_3CHO + HO_2 + HO_2 + CO_2$	$3.05 \times 10^{-15} \exp(-1900/T)$
* Very fast reaction step.	
n-butane chemistry	
$nC_4H_{10} + OH \rightarrow secC_4H_9 + H_2O$	$1.2 \times 10^{-11} \exp(-526/T)$
$sec\dot{C}_4H_9 + O_2 \rightarrow secC_4H_9O_2$	*
$NO + secC_4H_9O_2 \rightarrow secC_4H_9\dot{O} + NO_2$	3.0×10^{-12}
$secC_4H_9\dot{O} + O_2 \rightarrow HO_2 + CH_3COC_2H_5$	2.1×10^{-16}
$secC_4H_9\dot{O} \rightarrow CH_3CHO + \dot{C}_2H_5$	1.2×10^{3}
$CH_3COC_2H_5 + h\nu \rightarrow \dot{C}_2H_5 + CH_3CO$	6.5×10^{-6}
$CH_3COC_2H_5 + OH \rightarrow CH_3COCHCH_3 + H_2O$	3.4×10^{-12}
$CH_3COCHCH_3 + O_2 \rightarrow CH_3COCH\dot{O}_2CH_3$	*
$CH_3COCH\dot{O}_2CH_3 + NO \rightarrow CH_3COCH\dot{O}CH_3 + NO_2$	3.0×10^{-12}
$CH_3COCHOCH_3 + O_2 \rightarrow CH_3COCOCH_3 + HO_2$	*
$CH_3COCOCH_3 + h\nu \rightarrow CH_3\dot{C}O + CH_3CO$	1.4×10^{-3}
* Very fast reaction step.	
m-xylene chemistry	
	2.4×10^{-11} * * 3.1×10^{-12} *

TABLE 3. (continued)

$HCOCCH_3CHCHO + OH \rightarrow HCOCCH_3CHOHCHO$	2×10^{-11}
$HCOCCH_3CHOHCHO + O_2 \rightarrow HCOC\dot{O}_2CH_3CHOHCHO$	*
$HCOC\dot{O}_2CH_3CHOHCHO + NO \rightarrow HCOC\dot{O}CH_3CHOHCHO + NO_2$	3.1×10^{-12}
$HCOC\dot{O}CH_3CHOHCHO + O_2 \rightarrow CHOCHO + CH_3COCHO + HO_2$	*
$CHOCHO + h\nu \rightarrow HCHO + CO$	3.3×10^{-4}

* Very fast reaction step.

Process	Noon photolysis rate* (s^{-1})
Photochemical processes	
$O_3 + h\nu \rightarrow O(^1D) + O_2$	2.1×10^{-5}
$O_3 + h\nu + O + O_2$	3.7×10^{-4}
$NO_2 + h\nu \rightarrow NO + O$	5.7×10^{-3}
$NO_3 + h\nu \rightarrow NO_2 + O$	1.1×10^{-1}
$\rightarrow NO + O_2$	4.0×10^{-2}
$N_2O_5 + h\nu \rightarrow NO_2 + NO_3$	2.4×10^{-5}
$H_2O_2 + h\nu \rightarrow 2OH$	5.9×10^{-6}
$HNO_3 + h\nu \rightarrow NO_2 + OH$	3.3×10^{-7}
$HCHO + h\nu \rightarrow HO_2 + CO + HO_2$	2.1×10^{-5}
$\rightarrow H_2 + CO$	4.5×10^{-5}
$CH_3CHO + h\nu \rightarrow CH_3 + HO_2 + CO$	3.1×10^{-5}
$CH_3COC_2H_5 + h\nu \rightarrow CH_3CO\dot{O}_2 + C_2H_5\dot{O}_2$	6.5×10^{-6}
$CH_3COCOCH_3 + h\nu \rightarrow CH_3CO\dot{O}_2 + CH_3CO\dot{O}_2$	1.4×10^{-3}
$CH_3COCOH + h\nu \rightarrow CO + CH_3CHO$	6.5×10^{-4}
$HCOCOH + h\nu \rightarrow CO + HCHO$	3.3×10^{-4}
$CH_3O_2H + h\nu \rightarrow CH_3\dot{O} + OH$	4.4×10^{-6}

* At 50°N latitude, ground level.

et al. (1977), which assumes that scattering takes place only in a direction parallel to the direct beam of solar radiation.

Reflection by a cloud layer above the mixing layer is modeled by assuming that the cloud top and the base act as partially reflecting, homogeneous surfaces with a specified albedo, which is assumed to be the same for radiation from all directions and for all wavelengths (Derwent and Hov, 1979). In the model so far, the albedo is specified as 0.0, 0.3, or 0.6, according to the objectively analyzed relative humidity fields. Reflection at the surface is ignored.

Dissociation rate coefficients are calculated for every 5° latitude and every 15 min of the day. The total vertically integrated atmospheric O_3 column is adjusted to correspond to the season and latitude in accordance with the data given by Duetsch (1978). Points along a given trajectory are allocated J values through interpolation in time and space to the appropriate latitude and local time.

The chemical scheme chosen (Table 3) and the composition of the HC emissions adopted by no means represent a unique or formally optimized selection. Rather, the choice is based on a subjective judgment of what kind of chemical description is adequate to include in a model of long-range transport of oxidants based on the present understanding of how meteorological, chemical, and physical processes interact. Extending the chemical scheme to fill up any computer capacity available, both with respect to memory and to CPU time, is very simple. However, the important question is: To what extent would this improve the model performance, or would it just increase the complexity and intransparency of the model?

The modeling of diurnal behavior is very important in understanding the mechanisms underlying the long-range transport of oxidants (Hov et al., 1978a,b).

The concentrations assigned at the starting point of the 96-h trajectories can be important for the development along the trajectory, particularly when the photochemical activity is low and the chemical lifetime of both precursors and secondary pollutants is long. Ground removal is the ultimate removal mechanism for O_3, and where low deposition occurs, the O_3 lifetime is much longer than 4 days (Hov et al., 1978b). In such situations, even 4-day trajectories are insufficient to trace the history of an air mass. If the weather is fair at the starting point, the air masses arriving there may have already accumulated photochemically active pollution over a number of days. In such cases, air chemistry calculations are initiated up to 4 days before the start of the trajectory, depending on the length of the fair-weather period. The emission rates, the radiation values, and the meteorological parameters are then taken as averages over the 5 x 5 (25) grid squares surrounding the starting point of the trajectory. In this way, the chemical development along a model trajectory is made nearly independent of the initial conditions.

Air chemistry integration is started by using a set of concentrations corresponding to a very slightly polluted atmosphere, with the removal processes in equilibrium with NO_x and NMHC emissions averages near the Northern Hemisphere (2 x 10^{10} molecules/cm^2/s for NO_x and NMHC/NO_x = 1.25). The initial concentrations of the most important species obtained in this fashion are listed in Table 4.

Natural sources of HCs are not accounted for in the model. Separate model evaluations, indicate that natural HCs probably do not contribute significantly

TABLE 4. INITIAL CONCENTRATIONS

Species	Concentration (ppbv)	Species	Concentration
NO	0.14	NMHC (C)	3.38
NO_2	0.34	O_3	29.50
SO_2	1.27	HNO_3	0.19
SO_4	0.58	PAN	0.05

to oxidant formation on a regional scale in Europe (Derwent and Hov, 1980b; Hov et al., 1982). Natural sources of NO_x are thought to be small compared to the anthropogenic sources. Stratospheric O_3 or the O_3 concentrations in the free troposphere do not affect atmospheric boundary-layer chemistry as long as the upper boundary of the mixed layer is considered to be a material surface.

Emissions

Manmade NO_x and SO_2 emissions are mostly due to the combustion of fossil fuels. HC emissions arise mainly from incomplete combustion in motor vehicle engines and from evaporative losses of gasoline and solvents from storage and handling.

As a basis for the model calculations, emissions data for these gases were needed for a grid covering Europe. In principle, such data can be estimated, but the uncertainties are necessarily high. However, the degree of consistency obtained between the calculations based on a case study and the actual measurements (Eliassen et al., 1982) suggests that the estimates by and large reflect the actual emissions.

An inventory of European sulphur emissions was prepared in connection with EMEP (Dovland and Saltbones, 1979) giving the estimated annual (1978) emission in 150-km grid squares (the grid indicated in the lower right-hand corner of Figure 1).

The estimated total national emission figures are listed in Table 5. The uncertainty was 10% to 15% at best and considerably larger for many of the countries.

The estimates of national NO_x emissions in European OECD member countries (i.e., Austria, Belgium, Denmark, Finland, France, Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Luxembourg, The Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, and the United Kingdom) are based on information obtained from ongoing OECD work to evaluate possible photochemical oxidant control strategies. For the United Kingdom, however, the emissions data given by Apling et al. (1979) are used. For the remaining European countries, the emissions estimates are taken from Semb (1979), who has shown that, for OECD countries, national NO_x emissions are related to the total energy consumption, if deductions are made for hydroelectricity and nuclear electricity production and for noncombustion solid fuel used in the iron and steel industry. Data on energy consumption for all countries of the world are published by the UN Statistical Office. Thus, NO_x emissions for the remaining European countries have been estimated by assuming that the above-mentioned relationship is valid for these countries as well (Semb, private communication). The resulting estimated national emissions figures for NO_x are listed in Table 5. Chemically, NO_x is assumed to be emitted as NO. Uncertainties are likely to be larger than for the SO_2 emissions.

As a first approximation, NO_x emissions data in the 150-km grid were generated from the SO_2 emissions inventory by assuming that, for each country, the distribution of NO_x emissions on grid elements is identical to that for SO_2.

TABLE 5. ASSUMED ANNUAL EMISSIONS OF SO_2, NO_x, AND NMHC FOR ALL COUNTRIES IN EUROPE[a]

Country	Emissions		
	SO_2-S[b]	NO_x-NO_2[c]	NMHC[d]
Albania	50	10	10
Austria	215	275	280
Belgium	380	410	390
Bulgaria	500	240	240
Czechoslovakia	1,500	600	600
Denmark	228	240	220
Finland	270	200	200
France	1,800	1,650	2,000
German Democratic Republic	2,000	680	680
German Federal Republic	1,815	3,350	2,450
Greece	352	500	260
Hungary	750	220	220
Iceland	6	10	15
Ireland	87	90	105
Italy	2,200	1,550	1,750
Luxembourg	24	50	30
The Netherlands	240	700	600
Norway	75	110	170
Poland	1,500	1,000	1,000
Portugal	84	110	200
Romania	1,000	460	460
Spain	1,000	850	1,050
Sweden	275	260	380
Switzerland	58	160	260
Turkey	483	600	600
USSR (within grid)	8,100	5,000	5,000
United Kingdom	2,490	1,730	1,158
Yugoslavia	1,475	210	210
Remaining area within grid	256	50	50

[a] Emissions expressed in 10^3 tonnes.
[b] SO_2 measured as S.
[c] NO_x measured as NO_2.
[d] NMHCs measured by their total mass.

In grid elements where the sulphur emissions are thought to be anomalously high relative to the energy consumption, lower NO_x emissions were assumed. Due to the relatively large uncertainties of the NO_x emission allocated to the individual grid squares, a moving average over 3 x 3 (9) squares is employed in the model calculations.

The estimates of NMHC emissions are again based on information obtained from OECD (1982), except for the United Kingdom data, which are from Apling et al. (1979). According to these data, the ratio between national NMHC and NO_x emissions in OECD Europe varies between 0.5 and 1.82 (NMHC measured by total mass, NO_x measured as NO_2). For non-OECD European countries, the NMHC emissions were estimated to be roughly equal to the NO_x emissions.

The resulting NMHC emissions estimates are listed in Table 5. The uncertainty is thought to be considerably larger than that for SO_2 and may approach a factor of 2, particularly for non-OECD European countries.

The emission grid data for NMHC were generated by distributing the national emissions according to the sulphur emissions inventory. In areas where the concentrations from oil refineries and the petrochemical industry are high, increased NMHC emissions are assumed. As with NO_x emissions, a moving average over 3 x 3 grid squares is employed in the model calculations.

Mathematical Formulation

Based on the assumptions made above, the mass conservation equation determining the mass concentration c_i of species i can be written as:

$$\frac{Dc_i}{dt} = -\left(\frac{v_d}{h} + k_w\right) C_i + \frac{E_i}{h} + S_i$$

where D/dt = the Lagrangian (total) time derivative along a trajectory;

$v_d(x,y,t)$ = the dry deposition velocity, assumed to be variable for O_3;

$h(x,y,t)$ = the variable mixing height;

$k_w(x,y,t)$ = the wet deposition rate, active only during the rain (i.e., at a relative humidity >0.9%);

$E_i(x,y)$ = the direct emission of pollutant, in mass per unit area and time, from emissions inventories; and

S_i = chemical source or sink.

In general, S_i consists of terms of the type:

$$\pm k \prod_{j=1}^{N} C_j^{m_j} \text{ and } \pm n_j J_j(x,y,t) C_j,$$

which describe production or destruction by gas-phase reactions and photodissociation, respectively, where k is the reaction-rate coefficient, m_j is the order of the reaction with respect to the species number j, J_j is the dissociation rate, and n_j is a stoichiometric factor. The dissociation rate depends on the cloud cover parameterized by the relative humidity, season, latitude, local time, and the vertically integrated atmospheric O_3 column.

The first step in the integration procedure is to calculate the appropriate back trajectories from the analyzed wind fields. The second step is to convert the quantities v_d, h, k_w, E_i, and J_j, originally given as Eulerian fields, into Lagrangian information, i.e., as functions of transport time along the trajectories. These operations transform the mass conservation equation into an ordinary differential equation in time. Thus, the third step is to integrate the equation to obtain calculated instantaneous concentrations for the receptor points at the arrival times of the trajectories.

The mass conservation equations for all species in the model were integrated by using a quasi-steady-state approximation (QSSA) method, described in detail by Hesstvedt et al. (1978). This method is explicit and applies a fixed time step. When compared with Gear-type methods with automatic error control (Hesstvedt et al., 1978; Derwent and Hov, 1979), the method gave accurate predictions in a wide range of model calculations of atmospheric chemistry.

A time step of 15 min is applied. Comparisons with results obtained with shorter time steps showed that 5% is an upper limit for the computational error and that the sign of the error is usually distributed quite evenly between plus and minus.

The temporal resolution of the model is adjustable. Presently, instantaneous concentrations are calculated along 96-h trajectories with an arrival time every 6 h (0000, 0600, 1200, and 1800 GMT). The machine time required to compute 24-h O_3 levels on a CDC/CYBER 170/835 (i.e., four trajectories, each 96 h, at a single receptor) is 9.5 s. This includes the time it takes to compute the trajectories from the horizontal wind field. The dissociation rate coefficients are calculated separately. At midsummer, calculating 13 dissociation rate coefficients every 15 min of the day, on a grid corresponding to every 5° latitude, for albedos 0.0, 0.3, and 0.6, requires approximately 1000 s of CPU time on the same computer. These calculations are performed once and for all, however, and therefore contribute very little to the total consumption of computer time during the experimental stage of the model. Also, the dissociation rate coefficient calculations may be speeded up significantly if the expense of computer time is an important constraint on model work (Derwent and Hov, 1979).

A CASE STUDY

The model was tested against actual measured oxidant data in Southern Scandinavia, April 6-13, 1979 (Eliassen et al., 1982). A brief summary of the test is given here.

In south Norway a ground-based network of continuous O_3 recorders is operated every summer by the Norwegian Pollution Control Authority at Langesund (see Figure 1). Langesund is a coastal site that is not influenced by local pollution when the air masses come in from the sea. Ozone measurements made at Langesund and daily aerosol sulphate and SO_2 measurements made at Rorvik near Gothenburg were used to test the model. The latter station is run by the Swedish Water and Air Pollution Research Institute (IVL) and is part of the Swedish EMEP station network.

The study period was April 6-13, 1979. During this period, there were no frontal passages or rain over south Norway. The 96-h, 850-mbar, 1200-GMT trajectories shown in Figure 1 indicate easterly transport over distances up to 4,000 km. On April 13, the synoptic situation changed, resulting in transport from the southwest. On April 6, O_3 monitoring began. Maximum hourly concentrations of over 75 ppbv of O_3 were measured at Langesund from April 10-12 during the afternoon (see Figure 2), which strongly suggests that O_3 generated photochemically from pollution had accumulated in the atmospheric boundary layer.

The reference model was used to calculate O_3 concentrations at each of the four receptor points at Langesund. Figure 2 shows the calculated mean concentration and the standard deviation around the mean at 0000, 0600, 1200, and 1800 GMT during the selected time period. The largest standard deviations

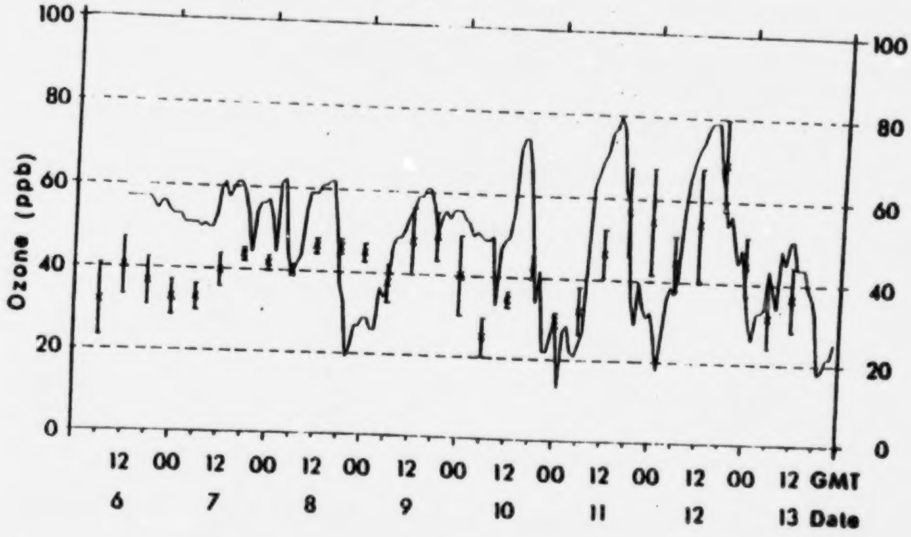


Figure 2. Mean and standard deviation of calculated O_3 concentrations at the four receptor points surrounding Langesund. The mean of the four values is denoted by X. The measured O_3 concentration is shown by a solid line.

are around 15 ppb, indicating that the calculated concentrations at the four receptor points are clearly different in some cases. This is due to the different trajectory paths and the pollutant emission fields, which exhibit large spatial variations. In the following discussion, all the O_3 model results presented are averages over the four receptor points around Langesund. These were averaged in order to smooth out variations in the calculations that are due to random errors in the wind field.

Figure 2 also shows the measured O_3 values at Langesund as a full line. In general, O_3 is underestimated during the selected period. The calculated O_3 peaks for April 10, 11, and 12 have increasing maximum values instead of recorded high peaks with a relatively constant maximum. The reduction of O_3 on April 13, associated with the shift in transport direction, is predicted by the model. In some cases the measured O_3 drops to a very low value at night. This may be partly due to suppression of vertical mixing by nighttime stabilization of the air near the ground, combined with dry deposition of O_3 (Garland and Derwent, 1979).

In a series of tests, the sensitivity of the calculated O_3 concentrations to variations in the initial pollutant concentrations, the O_3 deposition velocity, the advection wind, and the emission strength and composition was investigated. "Reasonable" variations in each of these quantities affected the calculations noticeably, suggesting that better knowledge might lead to improved calculations. The emissions and the deposition velocity for O_3 seemed to be the most important quantities for determining the O_3 concentration. The model calculations further showed that the production of O_3 from the primary pollutants emitted into the boundary layer can take several days, due to the inhibition of sunlight by cloud cover and to the chemical stability of some HCs.

As an example of the sensitivity studies, Figure 3 shows the results of a model run with zero emission outside Scandinavia. In this case, O_3 is seriously underestimated, and characteristic features in the measurements are not reproduced. This strongly suggests that the O_3 or its precursors to a large extent originate outside Scandinavia.

In order to shed further light on the transport process and the origin of the pollutants, we have studied the time development of the calculated pollutant concentrations in the air parcel arriving at one of the receptor points near Langesund at 1200 GMT on April 12. The trajectory of this air parcel is shown in Figure 1. The parts of the trajectory that pass over very large NMHC and NO_x emisisons (more than 10^{11} molecules cm^{-2} s^{-1}) are represented by a broken line. About 75 ppb of O_3 is measured at Langesund upon arrival of the trajectory. The calculated O_3 concentration in the air parcel at the time is also close to 75 ppb.

The time development of the O_3 and NMHC concentrations and the accumulated emission of NMHC into the air parcel are also shown in Figure 4. On the first day, the initial O_3 is slowly depleted by dry deposition until the air parcel encounters the large emissions in the Donbass area of the USSR, after about 24 h of transport. At night, the O_3 is further depleted by reacting with the emitted NO. There is no sunlight available so that O_3 can be produced from the emitted NMHC, which is therefore accumulated. The O_3 production on the second day is

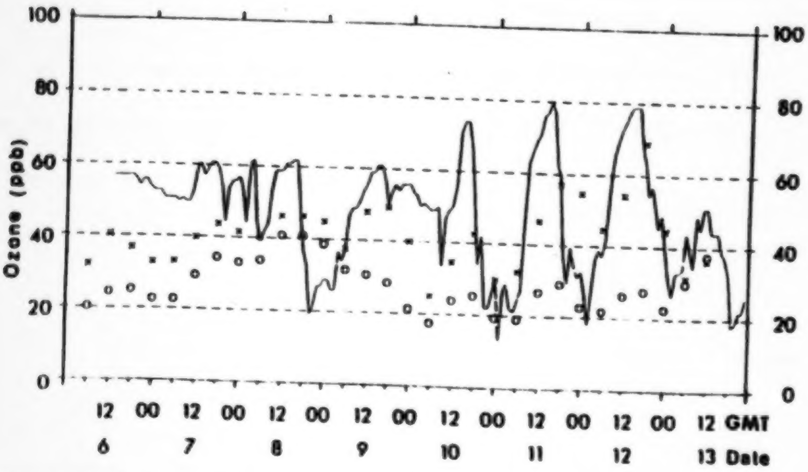


Figure 3. Sensitivity to emissions outside Scandinavia. The circles denote calculated O_3, with emissions of pollutants only from sources in Denmark, Norway, and Sweden. The reference model run is denoted by X, and measured O_3 concentrations are denoted by a solid line.

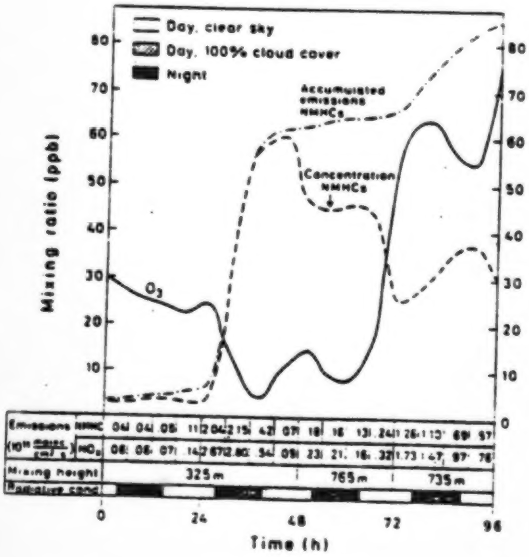


Figure 4. Time development of pollutant concentrations and emissions for the trajectory arriving near Langesund at 1200 GMT, April 12 (shown in Figure 1). The concentration of O_3 and NMHC and the accumulated emission of NMHC are shown as curves. The emission fluxes of NMHC and NO_x and the mixing height are shown in numbers below the curves. The radiative conditions are also indicated.

rather weak due to cloud cover, and only about 25% of the NMHC is consumed in the reactions. The third day is clear, and a considerable amount of O_3 is produced. This production is due to the emissions encountered by the air parcel about 1-1/2 days earlier. The high emissions encountered over Poland during the evening of the third day and the following night result in further O_3 production on the fourth day.

ACKNOWLEDGMENTS

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DISCUSSION

R. Lamb: In your presentation you said that the receptor-oriented model has an advantage over the source-oriented model because in the former case you can treat nonlinear systems because you have no overlapping of plumes. In other words, you are saying that the plumes overlap in the source-oriented models and that this creates a problem treating nonlinear chemistry.

In the case of the receptor-oriented model, the counterpart of the overlapping plumes is the diffusion of the material into the parcel? It seems that the problem does not disappear by using the receptor-oriented approach. If you allow any lateral mixing in the model, you have to compute the ambient concentrations of the species outside the parcel.

O. Hov: That is probably right. At the moment, it does not take into account lateral diffusion. However, neglecting diffusion is not a good approximation for oxidant studies in which you are actually computing hourly average values.

MODEL FOR THE REGIONAL TRANSPORT OF PHOTOCHEMICAL OXIDANTS AND THEIR PRECURSORS IN THE UNITED KINGDOM*

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INTRODUCTION

Extensive monitoring programs in Europe and the United States have established that O_3, PAN, and visibility-reducing aerosol concentrations may increase significantly during the summer over large areas in fair-weather episodes. These episodes are associated with air masses that remain over, or have passed through, industrialized or populated centers, and a significant, anthropogenic influence has been demonstrated in simultaneous observations made of elevated O_3 concentrations and concentrations of manmade species, such as trichlorofluoromethane (Cox et al., 1975) or acetylene (EPA, 1975).

Unlike pollutants in an urban area, primary and secondary pollutants accumulate at different time and space scales in a well-developed summertime high pressure situation. The time scale for oxidant production in an urban area is typically a few hours, and high levels of both primary (precursor) and secondary pollutants are already present on the first day of an episode. The horizontal scale for such situations is typically 10 to 100 km. In a summertime high pressure cell, however, accumulation occurs over a number of days, and quite uniform levels of pollutants, well-mixed throughout the atmospheric boundary layer, may be found over an area of 100 to 1,000 km. At some distance from the source regions, the precursors may be completely reacted, resulting in high levels of O_3, sulphate, and other secondary pollutants. In Europe, the long-range transport of oxidants (O_3, sulphate, nitrate) in connection with high pressure systems has been widely reported (Atkins et al., 1972; Cox et al., 1975; Apling et al., 1977; Guicherit and van Dop, 1977; Schjoldager et al., 1978).

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Theoretical investigations of the residence time of air parcels in a moving high pressure system show that air masses in a circular high pressure cell with a radius of approximately 1,000 km and moving at a constant speed of 5 m/s may spend up to a week in the system (Vukovich, 1979; Vukovich et al., 1977). If the system moves at slower speeds, the residence time may increase. Based on a chemical formulation involving nine emitted HCs, photochemical models of such episodes indicate that O_3 generated from continental precursor emissions may follow the mean air flow over the United Kingdom and remain at elevated levels for several days (Hov et al., 1978). Over rural areas, O_3 in excess of 100 ppbv (1 part per billion by volume = 2.4×10^{10} molecules/cm^3) may build up over a time scale of several days (Isaksen et al., 1978). The physical formulation of these models does not include vertical resolution; furthermore, the polluted air masses are assumed to be well mixed and represented by boxes. Rather simple assumptions are made about dilution and transport.

Derwent and Hov (1982) used this approach to investigate the potential for oxidant formation from United Kingdom emissions of HCs, NO_x, and SO_2, and to evaluate the possibility of long-range transport of secondary species generated during anticyclonic weather conditions. The following sections provide a description of the model used by Derwent and Hov, followed by a brief discussion of some of the results they obtained.

MODEL DESCRIPTION

Physical Model

The most densely populated and industrialized part of the United Kingdom was selected for study. A stationary model box with the horizontal dimensions of 450 km x 360 km was situated over South Central England. No horizontal or vertical grid was introduced; all emissions were assumed to be well mixed instantaneously throughout the model volume. In anticyclonic weather conditions, there is a suppression of vertical mixing of the atmosphere above a certain height, where stable layers are formed. The base of the stable layer may be as high as 2 or 3 km (Pasquill, 1974), but it is occasionally much lower.

In simulating the long-range transport of pollutants in anticyclonic weather conditions, the bulk of pollutants in the boundary layer are of interest, extending vertically to a potential mixing height. The actual mixing height, however, exhibits a marked diurnal variation, building up from a very small value at night to a potential value through the day (Pasquill, 1974).

Data from balloon ascents at Cardington in Bedfordshire, England, show that, when a capped layer exists, the mean inversion height at midday, averaged over the whole year, is 800 m. During the three summer months, the average value is 1,300 m. During periods of anticyclonic weather conditions, the nocturnal mixing height in rural areas is typically a few tens of meters.

The approach used to represent this situation in the model is as follows: The mixing height is kept constant at 1,300 m day and night. During the day, both primary and secondary species are deposited at rates corresponding to the respective deposition velocities. During the night, when a shallow, stable boundary layer is established, only primary species (i.e., those emitted) are assumed to be deposited. Secondary pollutants such as O_3 and PAN, which have been generated during the previous day(s), are assumed to have a zero deposition rate at night. Only a small fraction of the total boundary-layer column of secondary species is trapped underneath the nocturnal inversion. Although these compounds may be completely depleted in the shallow layer next to the ground during the night (Garland and Derwent, 1978), the influence on the total budget in the boundary layer is minimal (with a 40-m-deep nocturnal boundary layer, only 40/1300, or 3%, is depleted through deposition), and it was disregarded in the present model approach.

Horizontal homogeneity is an assumption that is justified only when the sources of the precursor emissions are evenly distributed spatially. Traffic and domestic emissions may satisfy this assumption rather well. In cases with large single sources, such as power stations and oil refineries, a realistic model picture should look more like an assembly of plumes, which eventually may interact with each other, rather than a volume in which all emissions interact all the time.

Assuming that complete mixing may occur throughout may therefore seem unrealistic. However, it is sufficient that the time scales of the various physical and chemical processes are represented correctly relative to each other. In the present model formulation, this means that vertical and horizontal mixing must occur faster than chemical development. It is well established that the time scale of oxidant generation is several hours or more. The assumption of instantaneous mixing can therefore be reduced to the assumption that complete mixing take less than a few hours to achieve. This assumption may be partially satisfied in photochemical episodes.

Mathematical Formulation

Each model species satisifes the continuity equation, written as:

$$\frac{dC}{dt} = P_e + P_{ch} - (L_{ch} + L_d)C \qquad (1)$$

where C = the concentration of the compound in question,

P_e = the emission term,

P_{ch} = chemical production and loss,

$L_{ch} \cdot C$ = chemical production and loss, and

$L_d \cdot C$ = the loss rate due to ground removal.

The emission rate is defined as:

$$P_e = \frac{\psi}{H} \qquad (2)$$

where ψ = the emission flux of the species in question, and

H = the mixing height.

$$L_dC = \frac{V_d}{H} C \qquad (3)$$

where V_d = the deposition velocity (see Table 1). SO_2 and NO_2 are assumed to be removed by dry deposition at night, and the secondary pollutants are assumed to be unaffected.

No transport term is included in Equation (1), indicating that there is no interaction with surrounding air masses. Thus, depending on the direction of the general mean air flow, the predicted oxidant generation may be overestimated or underestimated. In O_3 episodes, there is usually an easterly air flow over the United Kingdom, carrying high concentrations of precursors and secondary species from continental Europe (Cox et al., 1975; Apling et al., 1977).

TABLE 1. DEPOSITION VELOCITES

Species	V_d (cm/s)
O_3	0.6
SO_2	0.8
HNO_3	1.0
NO_2	0.1
PAN and homologs	0.2

Emissions

Emissions data for NO, SO_2, CO, and various HCs (40 species in all) were input into the model volume (see Table 2). The emissions of the various species were split into eight source categories: (1) petrol engine motor vehicle exhaust emissions, (2) diesel engine emissions, (3) petrol engine evaporative emissions, (4) stationary fuel combustion, (5) solvent use, (6) industrial processes, except petroleum industry, (7) petroleum industry, and (8) natural gas leakage. The emissions, which were calculated from the annual rates for 1975, were generally based on a combination of emission factors, statistics of total fuel consumption, or other relevant data. The detailed procedure and results are outlined elsewhere (Derwent and Hov, 1979), and the average United Kingdom emission fluxes for each species are given in Table 2. The emissions were not given any diurnal variation in the model because the emphasis of the study was on multiday features, not primarily on effects that occur on a time scale of a few hours.

Chemistry

The chemical formulation of the model includes approximately 145 intermediate products and end products in addition to the 40 emitted species. About 300 reactions are required to describe the degradation pathways. A detailed discussion of the scheme is given by Derwent and Hov (1980). In the present context, emphasis was placed on how the distribution of compounds within the main groups of species--sulphur, nitrogen, HCs, O_3--changes during the model run.

TABLE 2. AVERAGE UNITED KINGDOM EMISSION FLUXES

Species	Emission Flux (molecules/ cm/s)	Species	Emission Flux (molecules/ cm/s)
NO	2.81×10^{11}	CH_3CHO	4.94×10^{8}
SO_2	4.23×10^{11}	C_2H_5CHO	3.70×10^{8}
CO	2.94×10^{12}	C_3H_7CHO	1.51×10^{8}
		iC_3H_7CHO	6.79×10^{7}
CH_4	1.22×10^{12}	C_4H_9CHO	3.46×10^{7}
C_2H_6	4.07×10^{10}	CH_3COCH_3	3.48×10^{9}
C_3H_8	8.64×10^{9}	$CH_3COC_2H_5{}^2$	2.43×10^{9}
n-C_4H_{10}	2.19×10^{9}	methylpropylketone	1.24×10^{7}
i-C_4H_{10}	9.88×10^{9}	methyl-i-propylketone	1.24×10^{7}
n-C_5H_{12}	1.80×10^{10}	CH_3OH	1.24×10^{7}
i-C_5H_{12}	3.20×10^{10}	C_2H_5OH	1.75×10^{10}
C_2H_4	2.56×10^{10}	1-butene	3.30×10^{9}
C_3H_6	9.58×10^{9}	2-butene	4.43×10^{9}
C_2H_2	2.41×10^{10}	2-pentene	2.02×10^{9}
toluene	1.81×10^{10}	1-pentene	3.62×10^{9}
o-xylene	7.04×10^{9}	2-methyl-1-butene	3.12×10^{9}
m-xylene	7.04×10^{9}	3-methyl-1-butene	2.19×10^{9}
p-xylene	7.04×10^{9}	2-methyl-2-butene	5.04×10^{9}
ethyl benzene	7.04×10^{9}	butylene	1.24×10^{9}
HCHO	2.84×10^{9}	benzaldehyde	3.87×10^{8}

Ozone generation occurs by the well-known mechanism:

$$RO_2 \rightarrow RO + NO_2$$

$$NO_2 + h\nu \rightarrow NO + O$$

$$O + O_2 + M \rightarrow O_3 + M$$

$$\overline{RO_2 + O_2 \rightarrow RO + O_3}$$

where RO_2 is any peroxyl radical. The peroxyl radicals (organic or inorganic) are produced through the degradation of HCs. A hydroxyl radical is lost through the initial attack on a given HC molecule. Radical species are generated and regenerated at various stages in the HC decomposition chains and on various time scales. Secondary species such as aldehydes are formed; these are important free radical sources when photodissociated. Increased radical concentrations are an important feature of oxidant episodes. They favor rapid generation of secondary pollutants and shorten the time scale for precursor degradation.

Hydrocarbons are oxidized to CO and CO_2 by way of aldehydes, ketones, glyoxals, etc., as intermediates that are also eventually coverted into stable products.

Oxides of nitrogen are converted into secondary products such as HNO_3 and PAN and its homologs through reactions such as:

$$OH + NO_2 \rightarrow HNO_3$$

$$CH_3COO_2 + NO_2 \rightarrow PAN$$

$$C_2H_5COO_2 + NO_2 \rightarrow PPN$$

where the peroxyacetyl radicals, CH_3COO_2, are formed from acetaldehyde by:

$$OH + CH_3CHO \rightarrow CH_3COO_2.$$

PAN is thermally unstable (Cox and Roffey, 1977); that is,

$$PAN \rightarrow CH_3COO_2 + NO_2 \ (7.9 \times 10^{14} \exp(-12530/T))$$

and the same decomposition mechanism is assumed for all PAN homologs.

PAN and its homologs are also removed by ground deposition, which can be estimated by using Equation (3) and the data in Table 1. The loss of gaseous HNO_3 to the aerosol is considered to be slow, according to evidence reported by Brosset (1978), and is set to a rate corresponding to a characteristic time of 2 days (Derwent and Hov, 1979).

The chemical conversion of SO_2 to sulphate aerosol is assumed to take place through the following reactions:

$$OH + SO_2 \rightarrow HSO_3 \ 1.1 \times 10^{-12} \text{ (Calvert et al., 1978)}$$
$$CH_3O_2 + SO_2 \rightarrow SO_3 + CH_3O \ 5.3 \times 10^{-15} \text{ (Kan et al., 1979)}$$
$$HO_2 + SO_2 \rightarrow <1 \times 10^{-18} \text{ (Graham et al., 1979)}$$
$$O_2 + HSO_3 \rightarrow HSO_5$$
$$HSO_5 + NO \rightarrow NO_2 + HSO_4$$
$$HSO_4 + O_2 \ldots \rightarrow HO_2 + H_2SO_4$$
$$SO_3 + H_2O \rightarrow H_2SO_4$$

It should be noted that the recent data evaluation (NASA, 1981) recommends a value of less than 5×10^{-17} cm^3/molecule/s for $SO_2 + CH_3O_2$. The gas-phase reactions of SO_2 compete with dry deposition, which is estimated by using

Equation (3) and the deposition velocity given in Table 1. Droplet-phase mechanisms for SO_2 oxidation by $O_2.O_3$ and H_2O_2 were not considered in this study. No removal processes for nitrate or sulphate aerosol were included in the model.

Numerical Procedures

The system of differential equations describing the time evolution of the chemical species in the model have been solved using a quasi-steady-state approximation (QSSA) method (Hesstvedt et al., 1978). The accuracy of the results have been assessed by comparing runs made with the Harwell computer program FACSIMILE (Chance et al., 1977). This program employs a variable-order Gears method. It is highly suited to the integration of large, stiff systems of differential equations, and has an error limit of 0.1%. The agreement between the QSSA method and FACSIMILE was better than 1% for most species, and the required CPU time with QSSA was about one-third that for FACSIMILE. A typical run using FACSIMILE on the IBM 3081 required about 2 min of CPU time. The computational error limit on the results presented here is thus about 1%.

A fully diurnal sun corresponding to summer, 50°N, was modeled. The dissociation rates were calculated by using the scheme developed by Isaksen et al. (1977). A value for the diurnal variation in temperature (daily maximum of 25°C, daily mean of 17°C) and a value for relative humidity were also included. Relative humidity was given a maximum value close to 85% around dawn and a minimum of 45% around noon. The initial conditions were established by midnight on the first day by running the model from noon till midnight on day "zero", using the same set of model parameters specified for the main run.

RESULTS AND DISCUSSION

The potential for oxidant formation in a typical, fairly persistent high pressure cell was evaluated by integrating the model equations over a period of 4 days with emissions. The emission fluxes were then set to zero, and the model integration was continued for 3 days. The decay of the various species during this period allowed an assessment of their respective lifetimes and demonstrated the possibilities for long-range transport.

Ozone

Figure 1 shows how O_3, which was of primary interest, accumulated to reach 130 ppbv on the fourth day. A maximum in the net gas-phase generation of O_3 occurred on the second day, with gradually more and more being deposited due to the increased O_3 level. Even after emissions have been abolished, a substantial O_3 production continues through days 5 and 6, demonstrated by the widening divergence between the O_3 curve and the dotted line for deposition only and calculated by assuming zero production and loss only by ground removal. On the seventh day, the slopes of the two curves are similar, indicating that the decline in O_3 is then mainly due to deposition. The results shown in Figure 1 clearly demonstrate the long lifetime of O_3 in old, weakly polluted air masses. This supports the previous evidence on the long-range transport of O_3 (Cox et

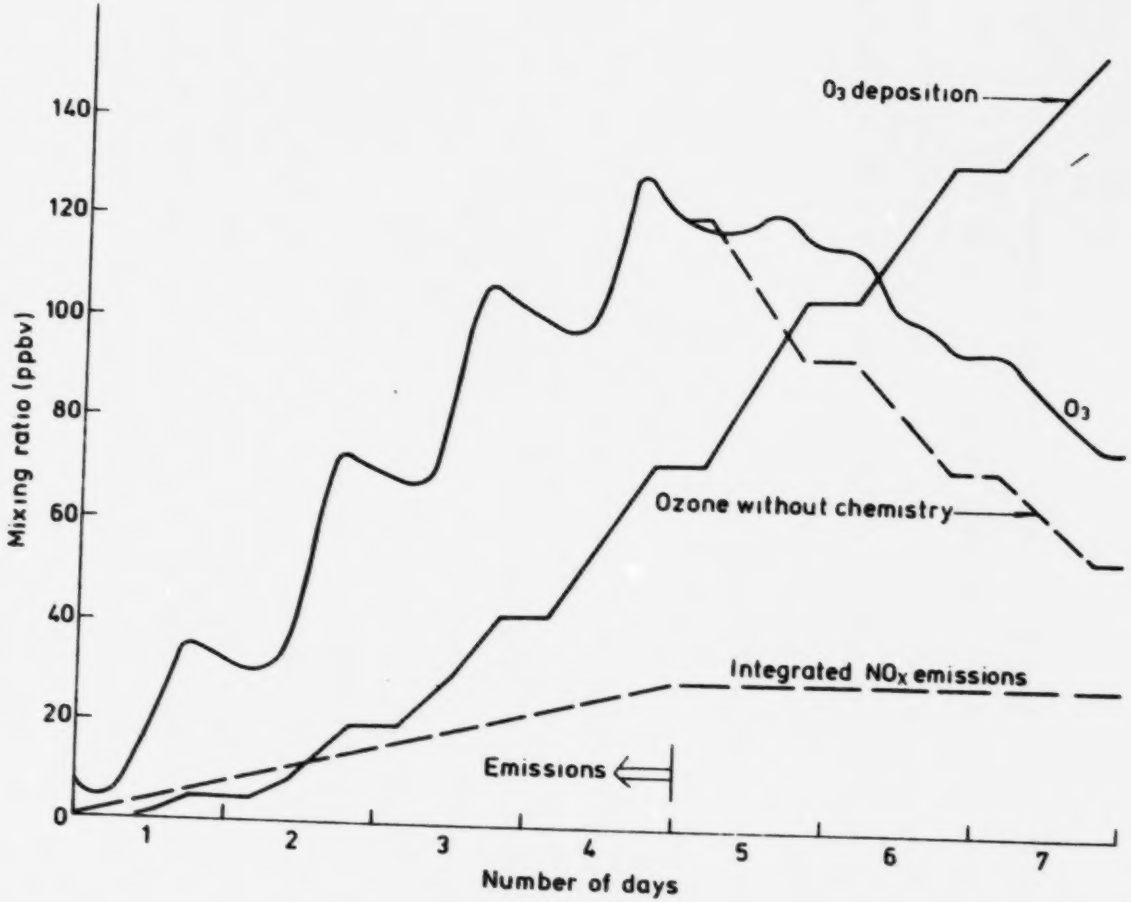

Figure 1. Development of gas phase and deposited O_3 with time. There are no emissions after day 4 (indicated by the vertical arrow). The integrated NO_x emissions are also shown, together with an analytically calculated curve for depletion through deposition only.

al., 1975; Apling et al., 1977; Hov et al., 1978). Such transport may be particularly important over sea surfaces, where the O_3 deposition is relatively slow.

Nitrogen Budget

Figure 2 shows that total gas-phase N species decrease with time, acompanied by an increase in nitrate aerosol and deposition of N species. Figure 3 shows the relative distribution of the gas-phase N species with time. Primary species (NO and NO_2) are replaced by secondary species (HNO_3, PANs), and it is evident that PAN and its homologs are important NO_2 carriers in aged air masses. Nitrate aerosol formation from HNO_3 is given a characteristic time of 2 days in the photochemical episode modeled here. Because the recycling of NO_x through dissociation of HNO_3 is slow, with a characteristic time of 10 days (noon dissociation rate), HNO_3 is an efficient sink for NO_x, while PAN and its homologs are only temporary sinks.

Hydrogen Budget

The HC budget is shown in Figures 4 and 5. There is a decline in total concentration from the fourth to the seventh day since TNMHC is defined here as:

$$\text{TNMHC} = \Sigma\text{olefins} + \Sigma\text{paraffins} + \Sigma\text{aldehydes} + \Sigma\text{aromatics} + \text{acetone} + \text{ketone} + \text{acetylene} + \Sigma\text{alcohols},$$

specifically excluding CO, PAN, and PAN homologs.

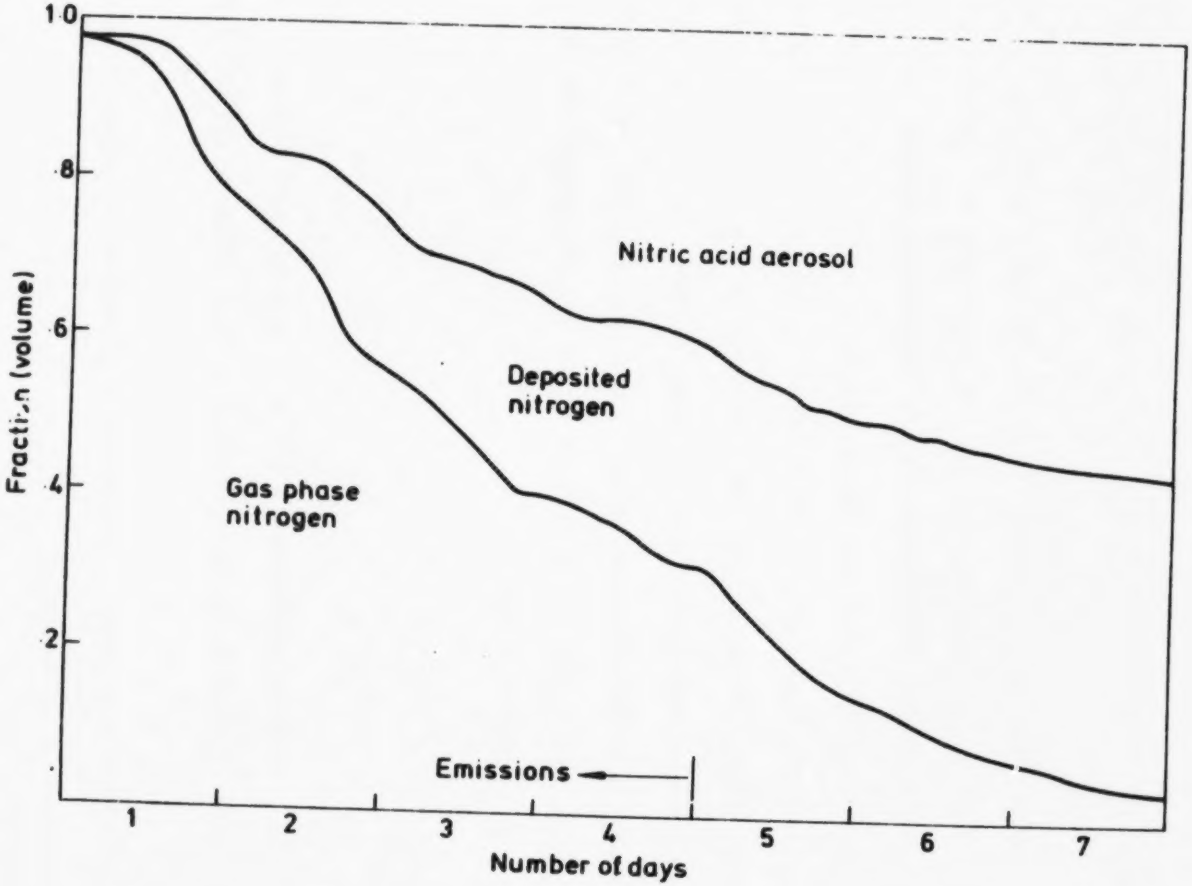

Figure 2. Relative distribution of nitrogen-containing species with time: gas phase, deposition, and aerosol. The relative fractions are on a volume (molecule) basis.

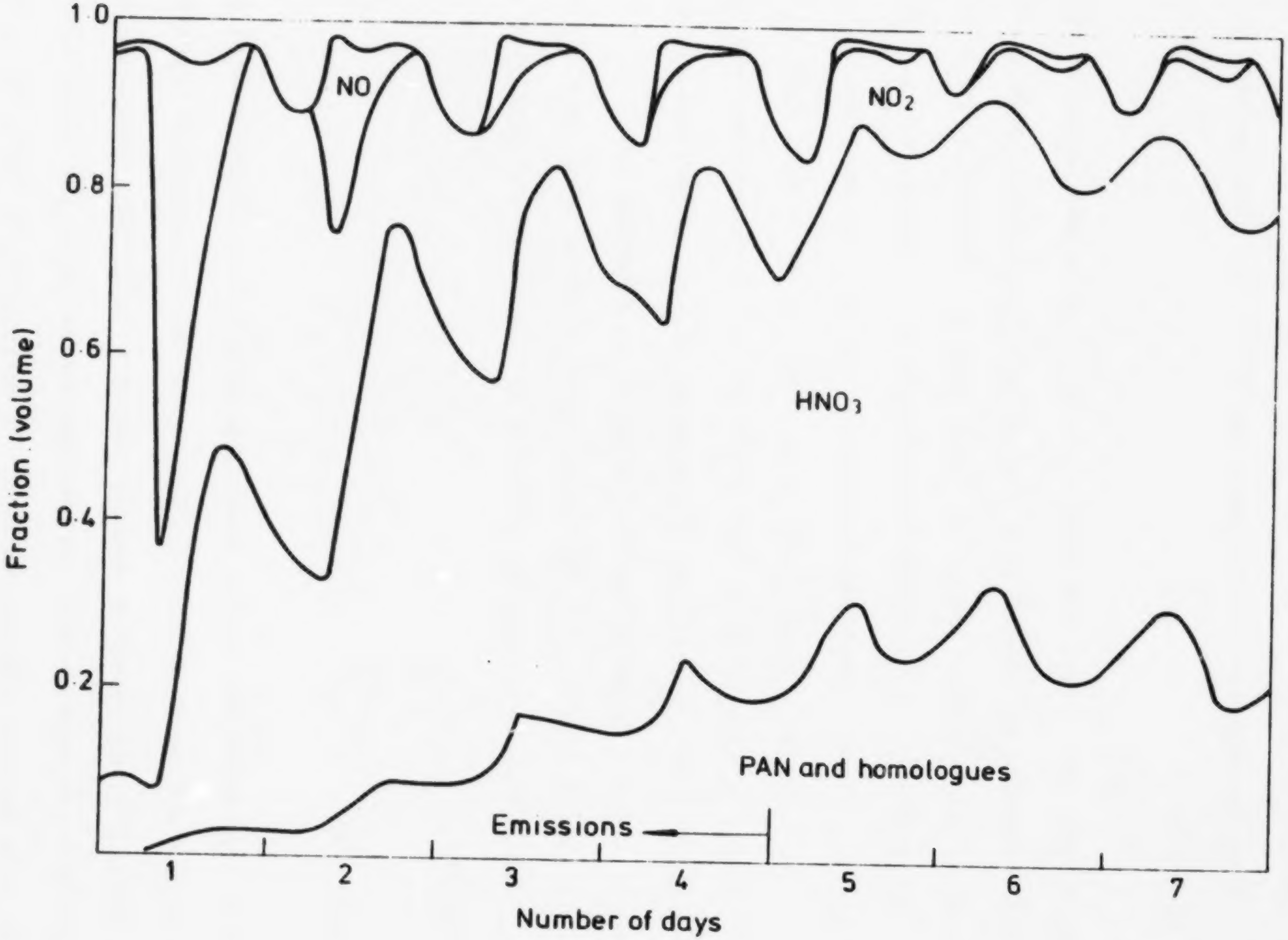

Figure 3. Relative distribution of gas-phase nitrogen-containing species.

Figure 4 shows there is a general increase in the relative fraction of species with low reactivity (paraffins, C_2H_2, CH_3COCH_3), while olefins and aromatic compounds virtually vanish. The relative distribution of the paraffins (Figure 5) shows the same trend: The lower reactivity fractions dominate more and more with time.

These calculations reveal that different mechanisms become important when the multiday formation of oxidants on a regional scale is compared with oxidant formation on an urban scale. During the first hours in a moderately polluted air mass, the behaviour of the oxidant formation is quite similar to that for an urban situation. After a few days, however, the less reactive HCs have accumulated and contribute a relatively larger share to the oxidant generation than the olefinic or aromatic HCs. Once emissions are abolished, the long-lived species assume an even greater role. In an aging, moderately polluted air mass, deposition is the most important constraint on the O_3 accumulation. Also,calculations have shown that the photochemical lifetime of O_3 in old air is very long, clearly demonstrating the potential of long-range transport from the United Kingdom.

Model Validation

Typical concentrations for the fourth and seventh days of integration of certain species are shown in Table 3. The fourth day concentrations represent overall maxima for low-reactivity species such as n-C_4H_{10}, HNO_3, and aerosols; other species reach approximately the same concentrations on the second and third day. The decline from the fourth to the seventh day reflects the chemical

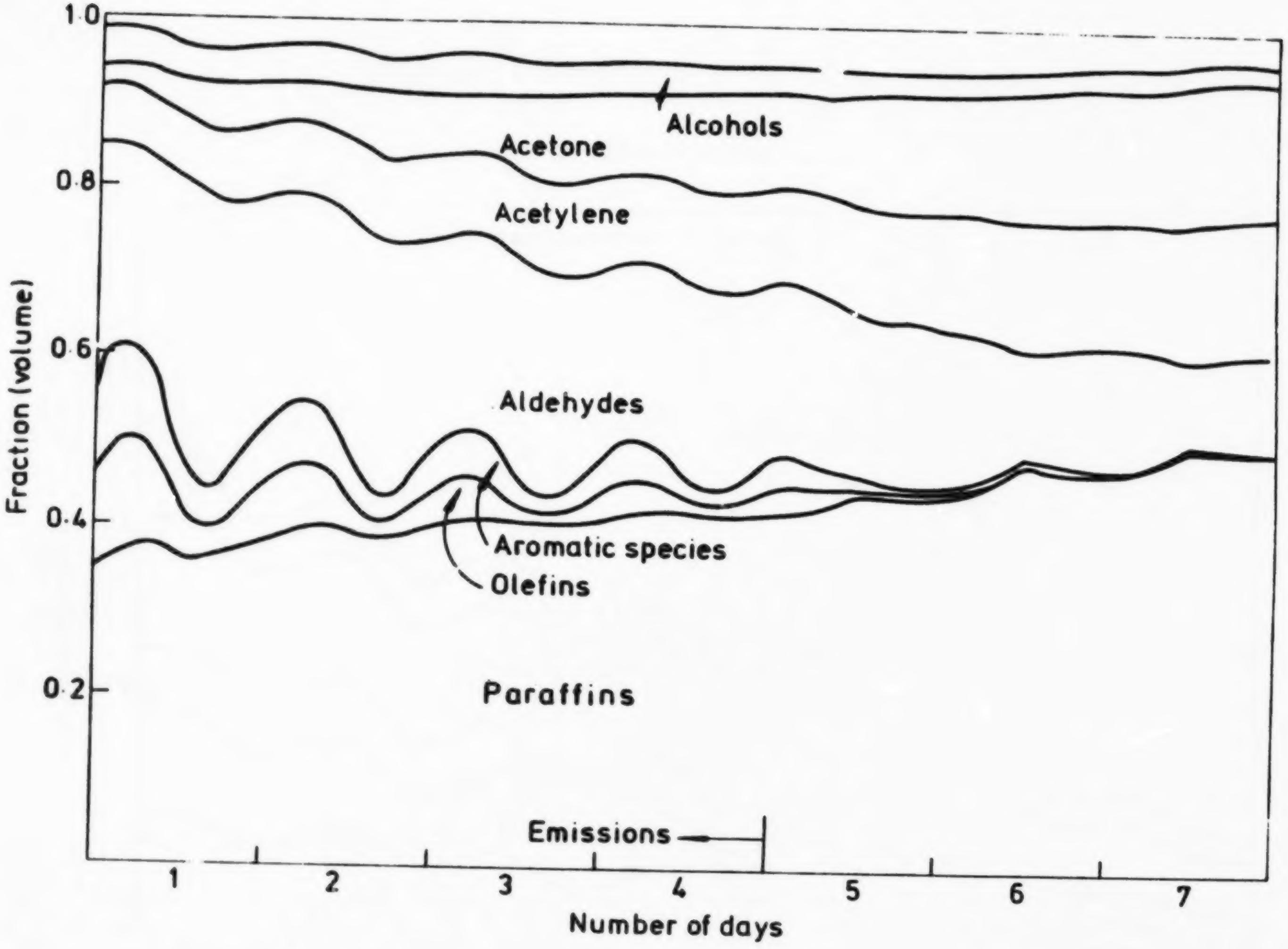

Figure 4. Relative distribution of the HC budget.

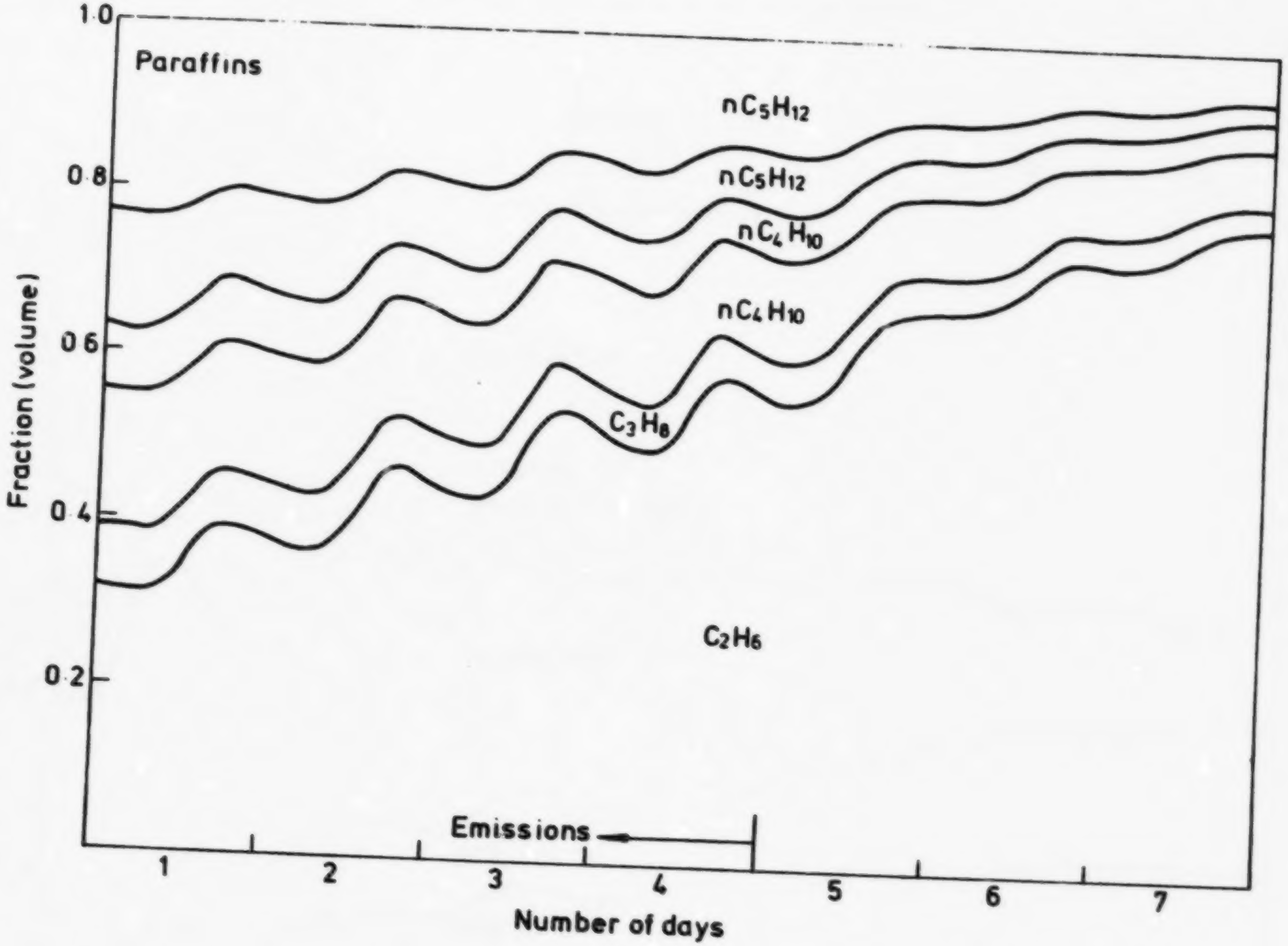

Figure 5. Relative distribution of the paraffins.

TABLE 3. COMPARISON OF MODEL-CALCULATED CONCENTRATIONS WITH OBSERVATIONS MADE IN RURAL SOUTHERN ENGLAND[a]

Species	Calculated		Measured	
	4th day	7th day	Concentration	Reference
OH	3.2×10^{-4}	1.3×10^{-4}	2.8×10^{-4}	Perner et al., 1976
			1.2×10^{-5} to 1.6×10^{-4}	Campbell et al., 1979
			1.3×10^{4} to 4.3×10^{-4}	Davis et al., 1979
HO_2	0.04	0.03		Cox et al., 1976
SO_2	1.3	2.8	7.5	Cox et al., 1976
$n\text{-}C_4H_{10}$	1.0	0.4	2.3	Cox et al., 1976
C_2H_4	0.5	0.01	2.3	Cox et al., 1976
HCHO	2.7	1.1		
CH_3CHO	1.0	0.4	2.7	Cox et al., 1976
NO_2	3.4	0.3	5.6	Cox et al., 1976
NO	0.6	0.04	2.1	Cox et al., 1976
PAN	1.9	0.5	2.0-8.9	Penkett et al., 1977
PPN	0.4	0.08	1/6 of PAN[b]	
HNO_3	6.2	1.4	1.7	Cox et al., 1976
O_3	130	96	<250	Apling et al., 1977
Nitric acid aerosol	34	47	15-30	Derwent and Hov 1979
Sulphate aerosol[c]	54	67	10-70	Atkins et al., 1973

[a] Concentrations in ppbv; aerosols in $\mu g/m^3$.

[b] The ratio between gas chromatogram peaks of PPN and PAN is fairly constant in all measurements and is usually around 1:6 to 1:8 (Penkett et al., 1975).

[c] In $\mu g/m^3$.

lifetimes of the various species. In general, primary species such as NO_x, SO_2, and reactive HC almost vanish, while O_3, PAN, and the PAN homologs remain at elevated levels.

Table 3 also shows measured concentrations from these trace species. These were determined at a rural site in southern England (Harwell), except for OH measurements made in Juelich, West Germany (Perner et al., 1976), in Tennessee, (Cambell et al., 1979), and in New Mexico (Davis et al., 1979). Ambient measurements made in the United Kingdom exist for only a limited number of species, and these are restricted to only a few sampling locations. In spite of the numerous assumptions and simplifications involved, the modeled calculations yielded concentrations that do at least appear to be realistic. Any refinement of the model will require improved validation data for precursors, intermediates, and secondary products.

SUMMARY

The results from this study indicate that photochemical oxidant formation in a persistent high pressure cell can be approximated by using a simple box-model formulation applied to United Kingdom precursor emissons data. The photochemical lifetime of O_3 in air that has left the source area is approximately 10 days, being determined principally by deposition. This demonstrates the potential for the long-range transport of O_3, particularly over surfaces such as water, where the deposition is inefficient. The calculations also show that reactive HCs are more important to photochemical oxidant generation on a regional scale than in an urban situation. PAN and its homologs are shown to be important carriers of photochemically active NO_x in aged air masses.

The simplified model described here is an initial attempt to simulate, on a regional basis, the general features of photochemical oxidation in the United Kingdom. It is not immediately applicable to the representation of oxidant levels experienced in the United Kingdom during typical episodes, when transport from continental Europe becomes important. The model runs can be regarded as an extreme case of a trajectory model, in which the air parcel is advected over a constant emissions field for a period of several days. Further work will require the construction of spatially and temporally resolved emissions inventories and the use of actual trajectories for past episodes to allow a more complete model assessment.

ACKNOWLEDGMENTS

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DISCUSSION

E. Runca: If I understand, your model is a one-dimensional model?

K. Brice: That is right.

E. Runca: There is almost no observation of meteorological factors affecting the development of photochemistry.

K. Brice: That is partially true. We are assuming a stationary box across southern England.

E. Runca: When you refer to a chemistry time scale of 10 h, exactly what do you mean, the time scale of the oxidants?

K. Brice: This is really based upon an assessment of an urban-plume-type approach where you look at the formation of, say O_3 or PAN, to some extent as well. However, I was interested in this major point for this discussion.

The time scale is really time taken as the maximum to be reached in that plume, and it is about 10 h. It perhaps actually balances between the formation rate and the k rate in that plume.

E. Runca: I think you would agree with me that the cycle of pollutants when they meet in the mixing layer are strongly affecting the sequence of reactions and the generation of secondary pollutants.

K. Brice: I agree with that point.

Unidentified Speaker: Your statment that, during the course of the multiday stagnation, the light HCs were actually increasing was unclear. I understand where the relative contribution would increase, but I was never aware that the total concentration of the HCs increased. Is that what you were saying?

K. Brice: No, the type of HC concentrations will obviously slightly decrease.

Unidentified Speaker: Was that just the relative contribution?

K. Brice: That was the relative contribution, right.

Unidentified Speaker: Also, I think you quoted Fred Vukovich from RTI. He indicated that the pollutants remain in the high-pressure system about 6 to 7 days, but that is the exception. I think ordinarily he is saying 3 to 4 days.

APPLICATION OF A REGIONAL OXIDANT MODEL TO THE NORTHEAST UNITED STATES*

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INTRODUCTION

A major concern over regional air quality is the long-range transport of O_3 and its precursors from upwind sources to rural areas. During several studies, O_3 concentrations exceeding the air quality standards have been observed over widespread areas beyond urban centers in the Eastern United States. Such incidences have also been reported elsewhere in the United States and in Western Europe. The movements of areas of high O_3 concentrations, frequently associated with increasing surface temperatures and decreasing visibility, generally correspond to movements of synoptic-scale high pressure systems. Many trajectory analyses seem to further implicate the long-distance transport of oxidants and oxidant precursors. For example, urban plumes emanating from St. Louis were tracked 160 km or more downwind. Air masses containing high O_3 concentrations over the Atlantic Ocean 250 km east of New York City were traced back to the metropolitan area. Similar observations were made in the Midwest and in California.

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*This report has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Current control strategies for photochemical oxidants are directed primarily at emission sources in the general vicinity of an urban area where excessive O_3 levels are observed. Thus, the possibility of O_3 or its precursors being transported from distant upwind sources certainly compounds emission control designs to reduce photochemial oxidants.

In order to arrive at an effective control strategy, contributions from local and distant sources must be quantified. Mathematical models have emerged as the most viable means for this assessment, based on the complex physical and chemical processes involved on this scale.

The objective of this paper is to describe an application and evaluation of a regional photochemical air quality model (RTM-III) developed by Systems Applications, Inc. Evolved from a regional transport/dispersion code, this model has undergone several iterations of improvements and modifications. The present application focuses on a regional air pollution episode recorded in July 1978 during the Sulfate Regional Experiment (SURE). A statistical evaluation of model predictions and observations has been completed.

MODEL EQUATIONS

The regional transport model evaluated in this paper is a descendant of a two-dimensional model developed by Liu and Reynolds (1983). This model, the oxidant version of the Regional Transport Model oxidant version (RTM-III), is based on the time-dependent, multispecies atmospheric diffusion equation for multiple layers:

$$\frac{\partial \bar{c}_i^{\,j}}{\partial t} + u\,\frac{\partial \bar{c}_i^{\,j}}{\partial x} + v\,\frac{\partial \bar{c}_i^{\,j}}{\partial y} = \frac{\partial}{\partial x}\left(K_x\,\frac{\partial \bar{c}_i^{\,j}}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y\,\frac{\partial \bar{c}_i^{\,j}}{\partial y}\right) \qquad (1)$$

$$+ E_i^j + F_i^j + R_i^j + Q_i^j - S_i^j \ ,$$

where the subscript i refers to pollutant species and the superscript j denotes the vertical layer. Three physical layers are used. The lower layer, designated by j = 1, consists of the surface layer and the mixed layer. The upper layer, designated by j = 2, is defined as the inversion layer. Other symbols have conventional definitions. An overbar denotes the layer average, u and v; K_x and K_y are the horizontal wind velocities and turbulent diffusivities in the x and y directions, respectively. The pollutant source and sink terms are denoted by Q_i^j and S_i^j, respectively. The source term Q_i^j consists of a gridded array of area sources whose emissions are dispersed into the lower layer, and emissions from major point sources enter the lower or upper layer as determined by the plume rise algorithm (Briggs, 1971). Entrainment/detrainment of pollutant mass due to the rise and fall of the layer boundary is represented by E_i^j, whereas the interfacial transport due to large-scale convergence or divergence is specified by F_i^j.

Parameterization of the Surface Layer

On a regional scale, surface depletion of pollutant mass has significant effects on the surface concentrations. The dry removal processes parameterized in the RTM-III consist of the diffusion of the pollutants from the mixed layer through the surface layer to the ground, followed by absorption or adsorption at the atmosphere/ground interface. The flux of pollutants of species i reaching the ground can be expressed in terms of the mixed layer concentration c by the expression:

$$f_i = \frac{\bar{c}}{I + (1/\gamma_i)} \qquad (2)$$

where

$$I = \frac{1}{\beta u_*} + \int_{z_0}^{h_s} \frac{\phi\left(\frac{z}{L}\right)}{k u_* z}\, dz \;, \qquad (3)$$

and

u_* = friction velocity,

k = the von Karman constant,

ϕ = the velocity profile function,

L = the Monin-Obukhov length,

h_s = the height of the surface layer,

z_0 = the roughness length, and

γ_i = the surface reaction rate constant that, for the first-order reaction, has dimensions of velocity.

Equation (2) can be derived by balancing the flux through the surface layer with the flux through the surface layer/viscous sublayer interface and the removal rate at the ground. The parameter β, analogous to the Stanton number in heat transfer. is the inverse of a dimensionless resistance for the viscous sublayer. Following Durran et al. (1979), β is given in the present study by:

$$\beta = \frac{u_*^{1/3}}{2.2} \; . \qquad (4)$$

This parameterization is more realistic than those commonly used in regional transport models, because it directly takes into account stability-dependent concentration profiles near the surface. The sink term in Equation (1) is related to the pollutant flux f_i and the mixed layer depth h by:

$$S_i^{\,j} = \begin{cases} f_i/^{h} & \text{if } j = 1 \\ 0 & \text{if } j = 2 \; . \end{cases} \qquad (5)$$

An equation for the pollutant loss per unit of area per unit of time can be written as (Benson, 1968):

$$f_i = \Upsilon_i \; C_i^{\,s} \qquad (6)$$

where f_i is the pollutant flux, $C_i^{\,s}$ is the surface concentration of species i, and Υ_i represents surface reaction rate constants. Combining this equation with Equation (2), we can thus determine the surface concentation $C_i^{\,s}$ from the average concentration in the mixed layer c by:

$$C_i^{\,1} = \frac{c}{1 + I \cdot \Upsilon_i} \; . \qquad (7)$$

Chemical Kinetic Mechanism

The chemical kinetic mechanism used in the regional-scale photochemical air quality model is based on the Carbon-Bond-II mechanism (Whitten, Killus, and Hogo, 1980). This mechanism is expressly carbon conservative, a feature of particular importance in situations involving extended transport and residence times. Hydrocarbon emissions used in the model are divided into five categories, according to the individual bonding structure of carbon atoms within each molecule. The carbon bond categories are: single-bonded carbon (PAR), ethylene (ETH), aromatic rings (ARO), and carbon-fast (OLE) and double-bonded carbonyl (CARB) groups.

The numerical solutions of the rate equations for chemical kinetic mechanisms generally encounter the stiffness problem. Stiff systems are defined as those with widely differing time constants. Classical methods for the solution of differential equations require a time step sufficiently small to avoid instability for the smallest time constant and may impose an enormous computational burden. Although a numerical algorithm based on the method of Gear (1971) can be used, the introduction of discontinuities into the forcing function of a Gear solution--such as changes in boundary conditions-can cause major inefficiencies in computation. These inefficiencies, together with the additional storage requirements for a high-order predictor-corrector method, make such a scheme unattractive in a grid model.

As an alternative, the invocation of the steady-state approximation can be used as an important tool for the numerical solution of chemical rate equations.

A transformation from differential equations to algebraic solutions allows the reduction of the number of species requiring numerical solutions. The species for which the steady-state approximation is valid are those with the smallest time constants. Thus, an elimination of these species greatly reduces the stiffness of the ordinary differential equations. The complexity of the algebraic solutions increases as more steady-state species that react with each other are included. For example, the implementation of the steady-state approximation in the Carbon-Bond-II mechanism requires the solution of a quintic polynomial for the selected radical species. One problem concerning the use of the steady-state approximation is that its use in effect removes that species from mass balance considerations; i.e., the steady-state expression assumes that $\dot{y} = 0$. If $\int \dot{y}dt$ is very small, then the removal of this species will not affect the overall mass conservation. If, however, $\int \dot{y}dt$ is sufficiently greater than zero, so as to affect the overall mass balance, then the steady-state assumption may produce invalid results.

This limitation has presented a potential problem in the use of the steady-state relationship for simulating major species such as NO, NO_2, and O_3. Generally, these three species are in dynamic equilibrium, with a time constant much faster than the rest of the nonradical chemical kinetic system. The dynamic equilibrium established is very close to:

$$K_3[O_3][NO] = K_1[NO_2] \tag{8}$$

Unfortunately, mass exchanges involving NO, NO_2, and O_3 are not small, and the conventional form of the steady-state approximation cannot be used. However, the simple steady-state values of NO, NO_2, and O_3 my be modified in terms of a correction factor:

$$\Delta_{ss} = -\frac{1}{2}\,[O_3] + [NO] + \frac{K_1}{K_3} + \frac{1}{2}\left\{\left([O_3] + [NO] + \frac{K_1}{K_3}\right)^2 - 4\left([NO][O_3] - \frac{K_1}{K_3}\,[NO_2]\right)\right\}^{1/2}, \qquad (9)$$

where

$$[NO_2]_{ss} = [NO_2] - \Delta_{ss}$$
$$[NO]_{ss} = [NO] + \Delta_{ss}$$
$$[O_3]_{ss} = [O_3] + \Delta_{ss}$$

If the state is variable, such as NO_x (=NO + NO_2) and unpaired oxygen atoms O_x (=O_3 + NO_2), the correction-factor equation becomes:

$$\Delta_{ss} = -\frac{1}{2}\left([O_x] + [NO_x] + \frac{K_1}{K_3}\right) + \frac{1}{2}\left\{\left([O_x] + [NO_x] + \frac{K_1}{K_3}\right)^2 - 4\,[NO_x]\,[O_x]\right\}^{1/2}, \qquad (10)$$

where

$$[NO_2]_{ss} = -\Delta_{ss}$$
$$[NO]_{ss} = NO_x + \Delta_{ss}$$
$$[O_3]_{ss} = O_x + \Delta_{ss}$$

Mass conservation is thus maintained for the two redefined species, NO_x and O_x. The steady-state calculation does not affect the quantity of these two species, it merely apportions them into the three molecular species--O_3, NO, and NO_2. In this scheme, NO-to-NO_2 conversions become the source of unpaired oxygen atoms.

This scheme has been tested against the Gear solution for a variety of cases. As might be expected, the approximation tends to break down when peroxyl radical concentrations are large relative to O_3 (i.e., when there are very high HC concentrations or very low NO_x concentrations). The former condition is not likely to be encountered, given the resolution of a regional-scale model. Under the latter condition, photochemical O_3 production is very slow relative to the background. Thus, the approximation appears to be reasonable for regional-scale applications.

APPLICATION OF THE MODEL TO THE NORTHEAST UNITED STATES

A rigorous model evaluation is predicated on a comprehensive data base that consists of meteorological data, an emissions inventory, and a spatially and temporally dense air quality monitoring network. The Regional Transport Model (RTM-III) described in the previous section has been used to simulate an episode of relatively high regional O_3 concentrations. The episode occurred over the eastern third of the United States during July 16 to 23, 1978. In addition to ample data coverage, the selection of this particular episode was also motivated by similar modeling exercises also focusing on the same period (Lavry et al., 1980; Niemann and Young, 1981; Bhumralkar et al., 1981; Stewart et al., 1983).

The modeling region selected for this study is almost identical to the EPRI/SURE grid, which is characterized by 80-km mesh squares defined over the eastern third of the United States. Dimensions of the grid subset are 2,080 km in the east-west direction by 1,840 km in the north-south direction. The grid resolution selected for the model simulation is 40 km x 40 km.

Meteorological Data

Wind velocities and mixing depths were derived from the National Weather Service (NWS) radiosonde network. The spatial resolution of the network is approximately 200 km, whereas temporal resolution is 12 h. Data from the monitoring network are spatially and temporally interpolated to generate the necessary wind field on the 80-km grid and at 3-h intervals, as required by the model.

For this modeling application, transport winds were defined as the layer-averaged wind velocity between the ground and 1,500 m. This transport wind was calculated for each radiosonde station and observation time. Linear interpolation was then employed to compute the transport winds at intermediate 3-h intervals. The wind velocity at the center of each model grid was then determined from an inverse-distance-squared interpolation of the irregularly spaced radiosonde data.

The procedures for determining the depth of the mixed layer are similar to those for determining the wind field. Following Benkley and Bass (1979), the mixed-layer depth h is determined at 3-h intervals from linear interpolation between a nighttime minimum h_{min} and a daytime maximum h_{max}. The minimum depth is determined from $h_{min} = 53|v|$, where $|v|$ is the magnitude of the layer-averaged wind. The maximum depth h_{max} is derived from the morning sounding at height where the potential temperature is equal to the maximum surface temperature of the following afternoon. Across the modeling region, a minimum value of 200 m was imposed on the mixed-layer depth.

Although the model has finer resolution than the radiosonde network, only those atmospheric features resolvable by the network can clearly influence the concentration pattern simulated by the model. It should also be noted that winds and mixed-layer depths are updated at 3-h intervals in a stepwise manner in the model. No temporal interpolation on the scale of an integration time step (i.e., approximately 20 min) has been attempted.

Air Quality Data

Air quality data used in this study consisted of ground-level O_3, NO, NO_2, SO_2, and sulfate concentration measurements, collected in the EPRI Sulfate Regional Experiment (EPRI/SURE). A detailed description of this data base can be found in Muller et al. (1982). The ground-level monitoring stations for SO_2 and sulfate consisted of 45 Class I and 9 Class II stations. The ground-level monitoring stations for O_3, NO, and NO_2 consisted of eight Class I stations operating continuously during the modeling period.

Emission Data

The EPRI/SURE program also established a detailed emissions inventory for the study region. Anthropogenic emissions used in this study were compiled in the following manner (Klemm and Brennan, 1981): Seasonal mean emission rates were derived for various pollutants in each of several source categories (i.e., electric power plants, major industries, commercial industries, home heating, and surface transportation). For each category, average temporal emission variations were calculated for eight 3-h periods, considering factors such as weekend/weekday and time of day. Emissions from all categories were then combined into a major point-source file (including stack parameters) and an area-source file aggregated into an 80 km x 80 km grid. Emission estimates prepared in this manner, derived from the National Emissions Data System (NEDS) of the U.S. Environmental Protection Agency, various state agencies, and the provinces of Ontario and Quebec, are current through July 1977.

The SURE emissions inventory consists of more than 3,000 point soures considered as major sources (>10,000 tons of SO_x per year). To treat this number of point sources in an economical manner, the emissions were aggregated into 498 point sources. This was accomplished by combining all the emissions from point sources with common plant codes and assigning the aggregated emissions to a source with the highest NO_x emissions rate.

Point and area source emission rates were available from the SURE emissions inventory for SO_2, sulfate, NO, NO_2, high-reactive HCs, medium-reactive HCs, and low-reactive HCs. The NO and NO_2 emissions were combined to form the NO_x

emission rate. The low-reactive HC emissions were low-reactive and nonreactive compounds and were not included in this study. Both the medium- and high-reactive HC emissions were composed of different reactive compounds, and were combined and split into Carbon-Bond-II mechanism compatible compounds as listed in Table 1. The total emission rates for the entire modeling region are divided by point and area source categories for the seven emitted species in Table 2. The spatial distributions of NO_x and reactive HC emission rates for the modeling region are displayed in Figures 1a and 1b.

Other Model Input Data

Exercise of the RTM-III requires additional information such as estimates of the photolysis rate constants and the dry deposition rates for O_3, NO, NO_2, SO_2, and SO_4. The photolysis rate constant was diurnally and spatially varied as a function of the solar zenith angle.

The parameterization of dry deposition rates requires a characterization of the underlying surface. This is needed for estimating both the diffusion toward the surface and the absorption rate. Table 3 lists the representative surface roughness and the deposition velocities used for each type of surface encountered in the model simulation. The geographical distribution of different surface categories over the modeling region is illustrated in Figure 2.

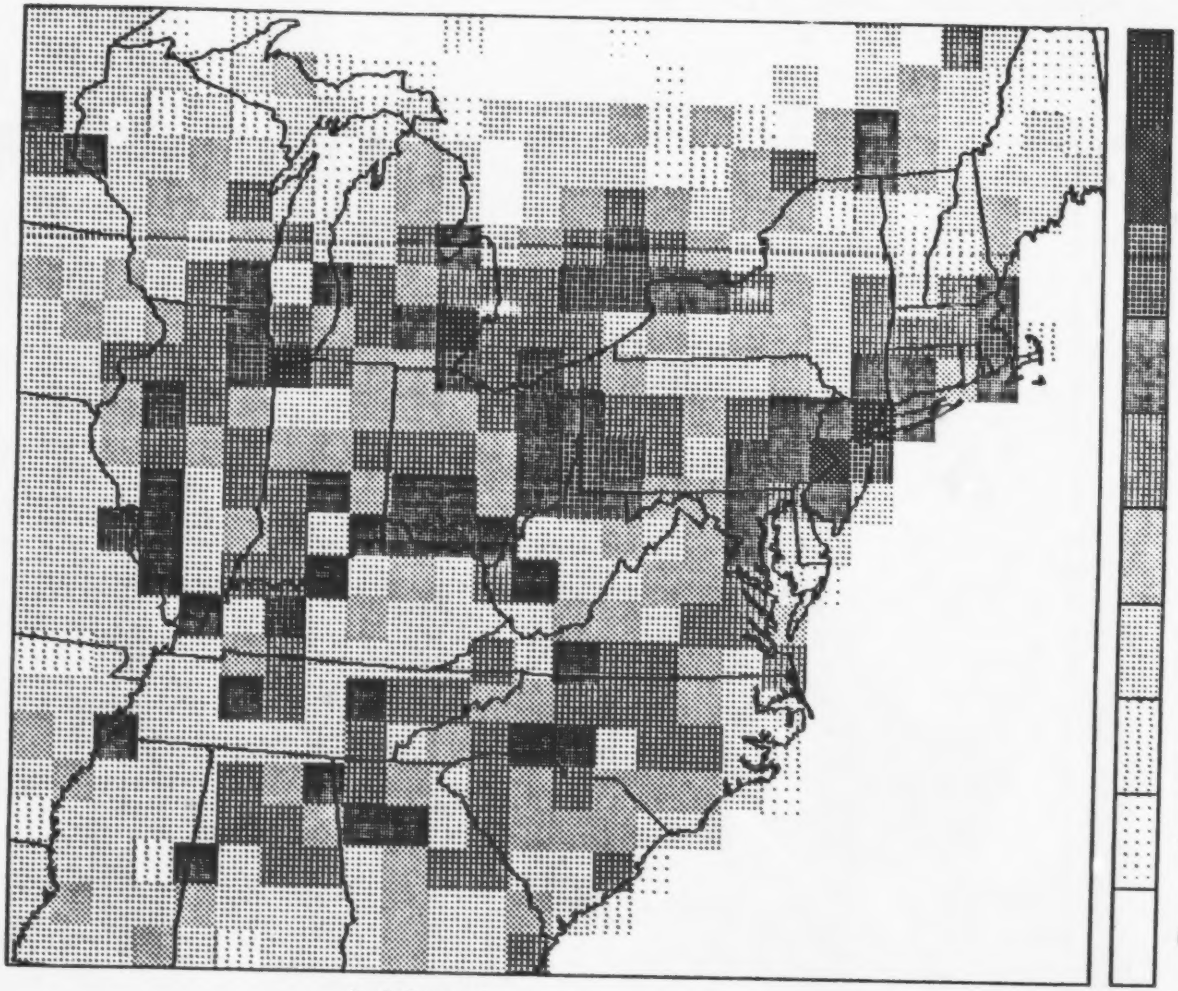
TABLE 1. HYDROCARBON SPLIT USED FOR HYDROCARBON EMISSIONS[a]

Compound	Normalized Fraction (%)
ETH	5
OLE	3
ARO	22
PAR	65
CARB	5
Total	100

[a]From Killus and Whitten, 1981.

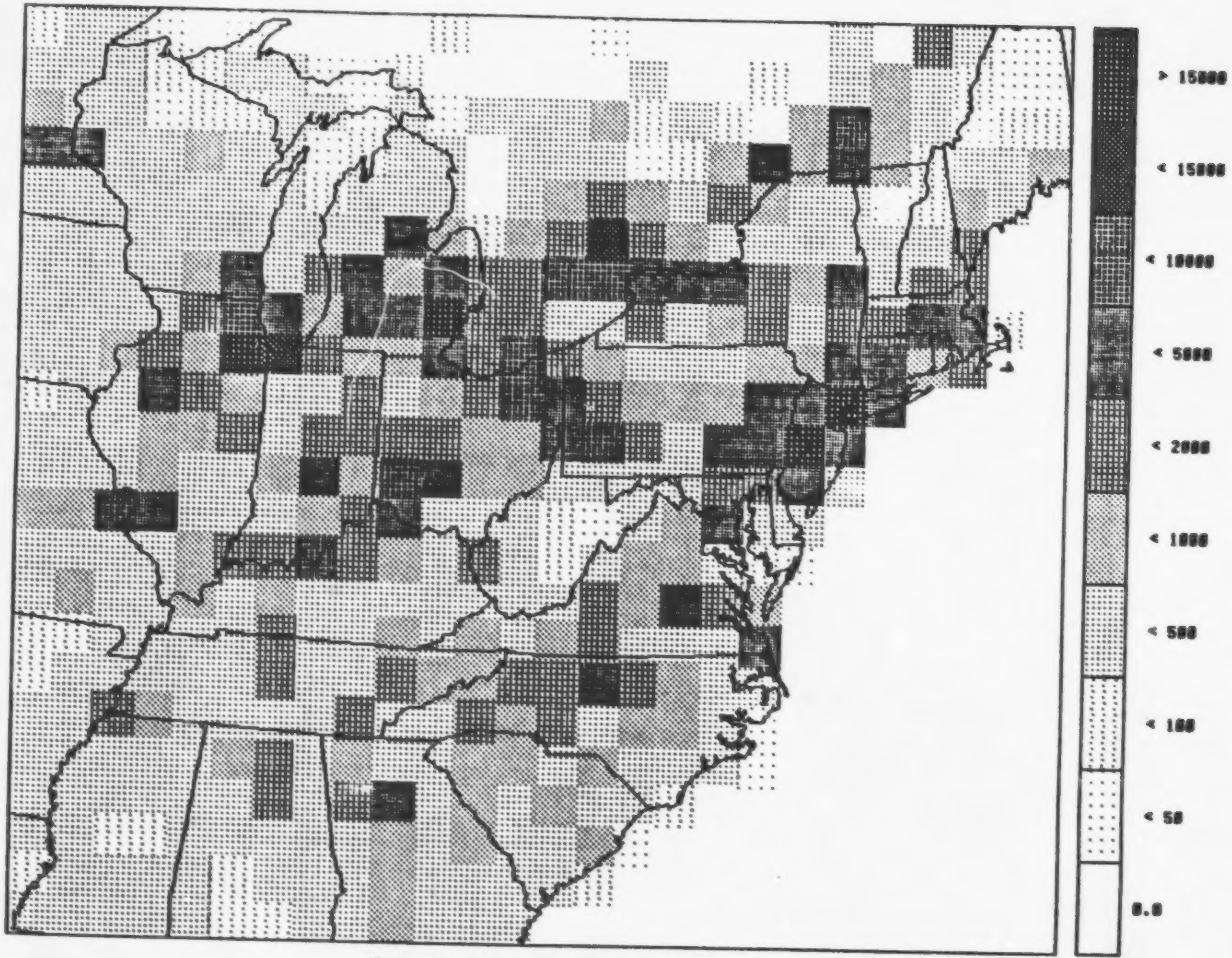
TABLE 2. TOTAL EMISSION RATE FOR THE MODELING REGION (tons/day)

Category	NO_x	ETH	PAR	CARB	ARO	SO_2	Sulfate
Major point sources	18,180	193	2,510	309	850	61,880	1,665
Area sources	34,207	2,020	26,255	3,231	8,887	20,633	1,021
Total	52,387	2,213	28,765	3,540	9,737	82,513	2,686


> 15000
< 15000
< 10000
< 5000
< 2000
< 1000
< 500
< 100
< 50
0

(a) NO_x (as NO_2)

Figure 1a. Total gridded emission rates (g/s): NO_x (as NO_2).


> 15000
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< 5000
< 2000
< 1000
< 500
< 100
< 50
0.0

(b) Reactive Hydrocarbon (as methane)

Figure 1b. Total gridded emission rates (g/s): Reactive HC (as methane).

TABLE 3. SURFACE CHARACTERIZATION BY TERRAIN TYPE

Terrain Type	Percentage of Modeling Region	Surface Roughness[a] (cm)	Deposition Velocity (cm/s)[b]					
			O_3	NO	NO_2	CO	SO_2	$SO_4^=$
Cropland and pasture	4.7	20	1.20	0.1	1.2	0.0006	0.70	0.20
Cropland, woodland, and grazing land	51.8	30	1.20	0.1	1.2	0.0025	1.00	0.20
Grazed forest and woodland	2.0	90	1.10	0.1	1.4	0.0014	1.20	0.40
Ungrazed forest and woodland	14.4	100	1.48	0.1	1.5	0.0045	1.50	0.50
Swamp	1.0	20	1.00	0.1	1.0	0.0008	0.14	0.35
Metropolitan city	0.2	100	2.00	0.1	1.8	0.0050	1.50	0.30
Lake or ocean	25.9	0.01	0.10	0.1	0.6	0.0005	1.10	0.50

[a] From Sheih et al., 1979.

[b] From McMahon and Denison, 1979.

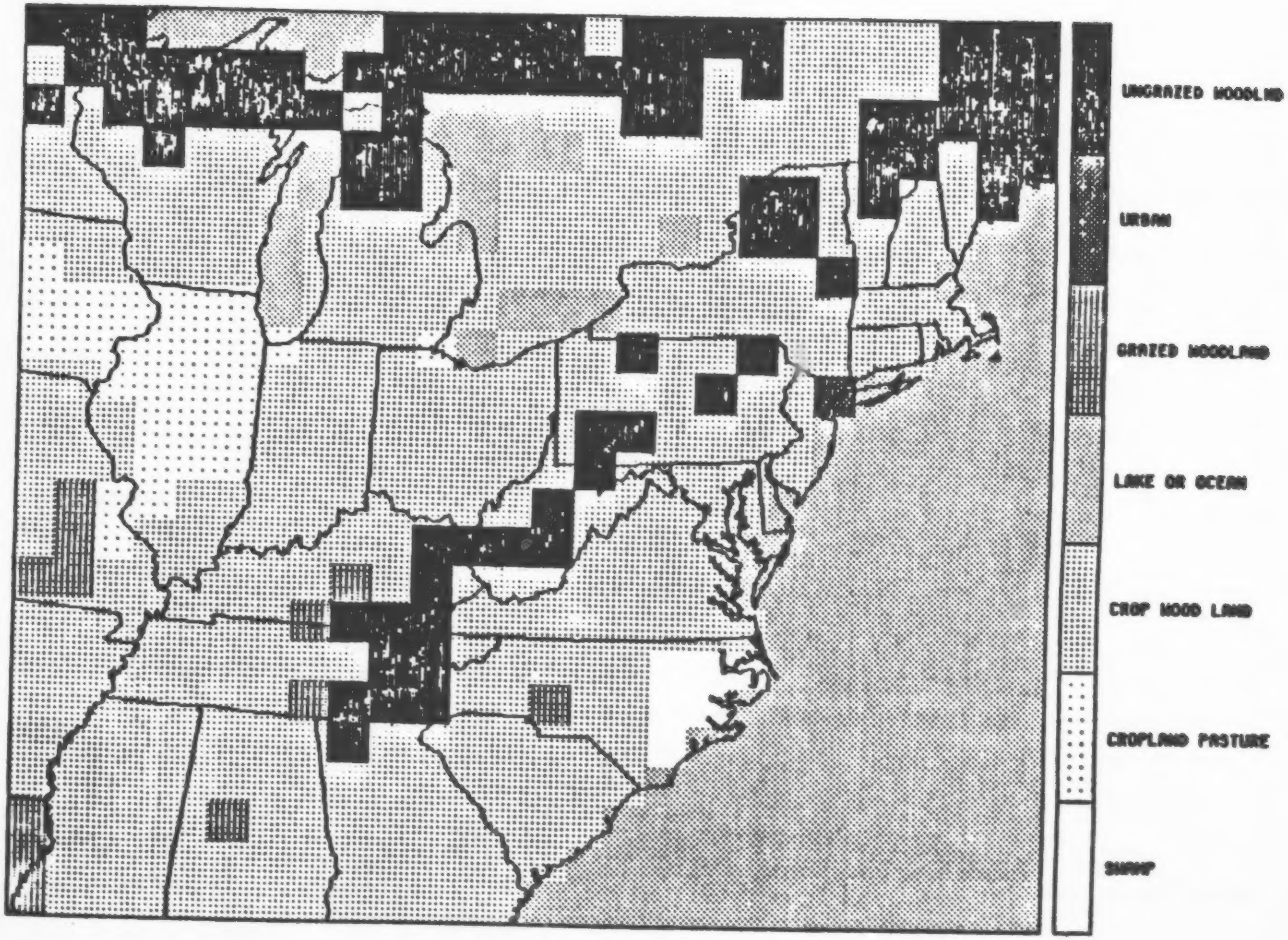
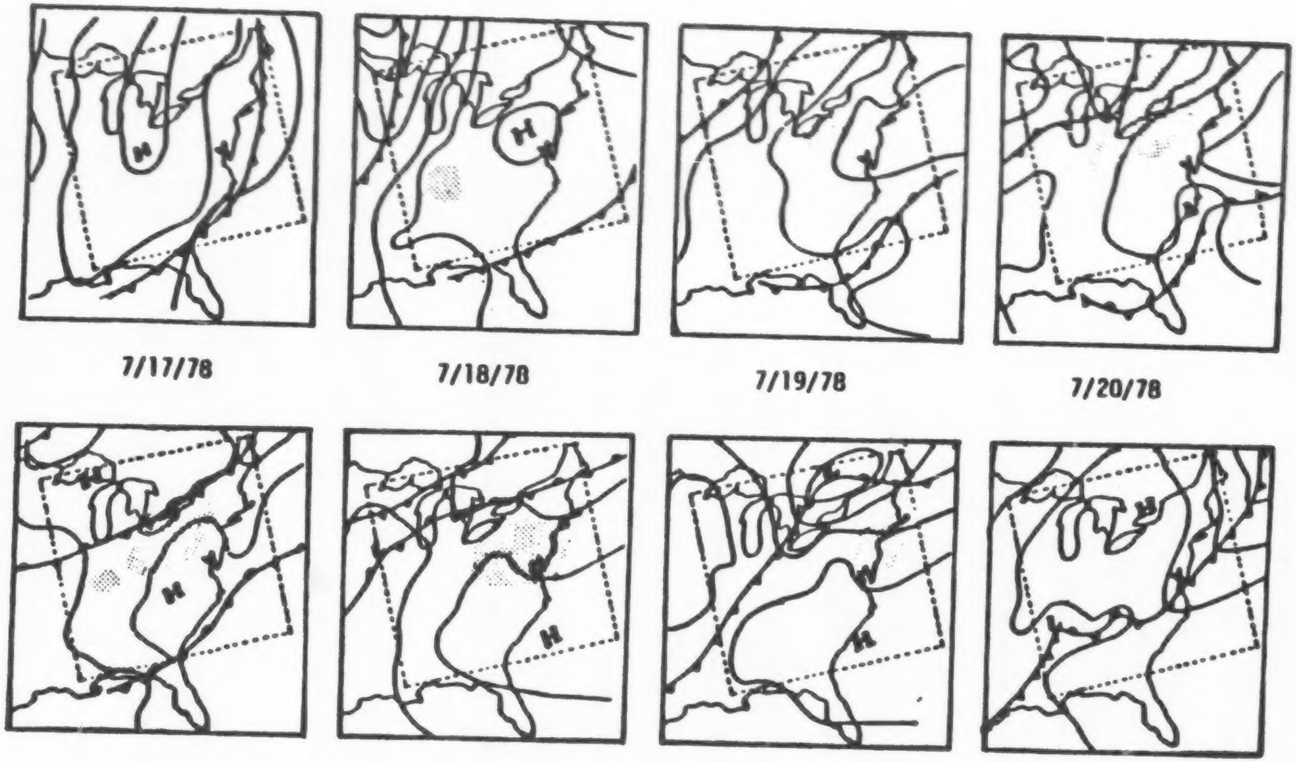

UNGRAZED WOODLND
URBAN
GRAZED WOODLAND
LAKE OR OCEAN
CROP WOOD LAND
CROPLAND PASTURE
SWAMP

Figure 2. Land use pattern in modeling region.

MODEL EXERCISES FOR THE JULY 1978 EPISODE

RTM-III was exercised for a period spanning eight consecutive days (July 16-23, 1978), the same period analvzed by Stewart et al. (1983) using the SO_2/sulfate version of the Regional Transport Model (RTM-II). Detailed discussions on the prescription of the meteorological input to the model can be found in Stewart et al. (1983). A brief synopsis of the 8-day sulfate episode selected for model evaluation is presented in Figure 3. The surface weather maps are representative patterns for 1200 GMT (0700 EST) of each day. Prior to July 17, 1978, a ridge of a high pressure system extended southward behind an advancing cold front. During July 18 and 19, 1978, the cold front slowly moved offshore, and the surface high pressure intensified to 1,020 mbar. The high pressure system drifted southward and expanded over the northern Atlantic Ocean. The eastern and central states remained under the influence of the high pressure system, which merged over the next few days with the Burmudian subtropical high. During this period, air flow at 500 mbar exhibited a zonal-oriented weak ridge that gradually evolved into a closed circulation over the eastern states.

Toward the northern and western portions of the modeling region, a cold front advanced and became stationary while the high pressure system to the east was strengthening. A low pressure system formed on this front over Iowa during the morning of July 22, 1978. This low, with its accompanying cold front, advanced across the modeling region late on July 23, 1978, displacing the high pressure system.



7/17/78 7/18/78 7/19/78 7/20/78

7/21/78 7/22/78 7/23/78 7/24/78

Figure 3. Sequence of surface synoptic maps during the sulfate episode with modeling region outlined (sulfate concentrations exceeding 30 $\mu g/m^3$ are indicated by shading).

During July 17 and 18, 1978, winds in the lowest 1500 m were generally light and northerly to the east and southeasterly to the southwest of the high pressure system. The onset of the closed circulation at 500 mbar on July 21, 1978, coincided with an increase in southwesterly winds northwest of the surface high. Prior to July 20, 1978, high sulfate levels were associated with stagnant or light wind conditions. A ducting situation developed after July 20, 1978, with the elevated sulfate region becoming elongated in the direction of the wind and gradually being transported eastward across southern New England and out to sea.

To ensure that the pollutant mass within the modeling domain reaches a quasi-steady state before evaluation, the model was exercised for 48 h prior to July 16, 1978. Initial concentrations and boundary values for the nine modeled species were held constant for the entire simulation and are listed in Table 4.

Tropospheric background concentrations of O_3 have been studied by numerous investigators (Jung, 1963; Aldaz, 1967; Ripperton and Worth, 1969; Rasmussen, 1975; Rutheir et al., 1980) and are established as 0.02 to 0.06 ppm. This range is exceeded only under unusual conditions involving either stratospheric intrusion episodes or anthropogenic contamination. For this modeling study, the middle range of concentrations (0.04 ppm) was used for background O_3. The modeling region is downwind of numerous anthropogenic source areas, so the boundary conditions may be affected by upwind sources.

Background concentrations of NO_x are not precisely established (Singh et al., 1980). However, they are known to be low, on the order of a few parts

TABLE 4. INITIAL AND BOUNDARY CONDITIONS USED BY RTM-III[a]

Species	Initial Condition	Boundary Conditions: North, South and Western	Boundary Conditions: Eastern
NO_x	1	1	1
O_x	40	40	30
ETH	1	1	0.1
PAR	35	35	35
CARB	15	15	0.1
ARO	0.8	0.8	0.1
PAN	0.1	0.1	0.1
SO_2	2	2	1
$SO_4^=$	2	2	1.5

[a] All concentrations in ppb except $SO_4^=$, which are in $\mu g/m^3$.

per billion at most. For the present model application, the value is set at 1 ppb. Use of this value may result in the generation of 10 to 20 ppb O_3 when combined with a background of reactive HCs.

On the basis of an extensive review of measurements, the total background concentration for HCs in the model is set at approximately 0.05 ppmC. This seemingly low concentration results in a HC loading of 4.7 x 10^4 metric tons within the model. This is only slightly smaller than the daily anthropogenic HC emission for the entire United States, about 6 x 10^4 metric tons in 1973 (EPA, 1976). The source of this HC background is not well understood. Certainly, the long-range transport from anthropogenic sources is a factor, along with natural geogenic and biogenic sources. Since model results are sensitive to this reactive background, a regional-scale modeling study will require a careful assessment of the distribution and relative impact of the background sources.

EVALUATION OF MODEL PREDICTIONS

Hourly predictions of various pollutants species (O_3, NO, NO_2, SO_2, and $SO_4^=$) from the RTM-III were compared with the corresponding observations. Statistical comparison of predictions and observations for the 8-day episode are summarized in Table 5. Reasonable agreement between the predicted and observed O_3 concentrations can be seen from the small average residual and a high correlation coefficient of 0.7. A scatter plot for the predicted and observed

TABLE 5. SUMMARY OF STATISTICS FOR PREDICTIONS/OBSERVATIONS[a]

Species	Averaging Time	Sample Size	Average Observation	Average Bias[b]	Average Absolute Residual	Correlation Coefficient
O_3	Hourly	1,508	49	0 ± 1.5	17	0.70
NO	Hourly	679	4	4 ± 0.4	4	0.07
NO_2	Hourly	662	9	7 ± 0.5	7	0.00
SO_2	3-h	2,838	26	2 ± 1.5	19	0.24
$SO_4^=$	24-h	362	16	-7 ± 1.5	9	0.77

[a] Concentrations in ppb except $SO_4^=$, which is in μg/m3.
[b] At 95% confidence intervals.

O_3 concentrations is shown in Figure 4. A comparison of the predicted and observed $SO_4^=$ concentrations is equally favorable with a correlation coefficient of 0.77, comparable to an identical study using the SO_2/sulfate version (RTM-II) reported by Stewart et al. (1983).

Comparisons of predictions and observations of NO_2 and primary pollutants such as NO and SO_2 are, however, quite disappointing. Contributions from local sources will certainly have significant effects on the model performance because of the limited spatial resolution of the model. The poor NO_2 predictions, a

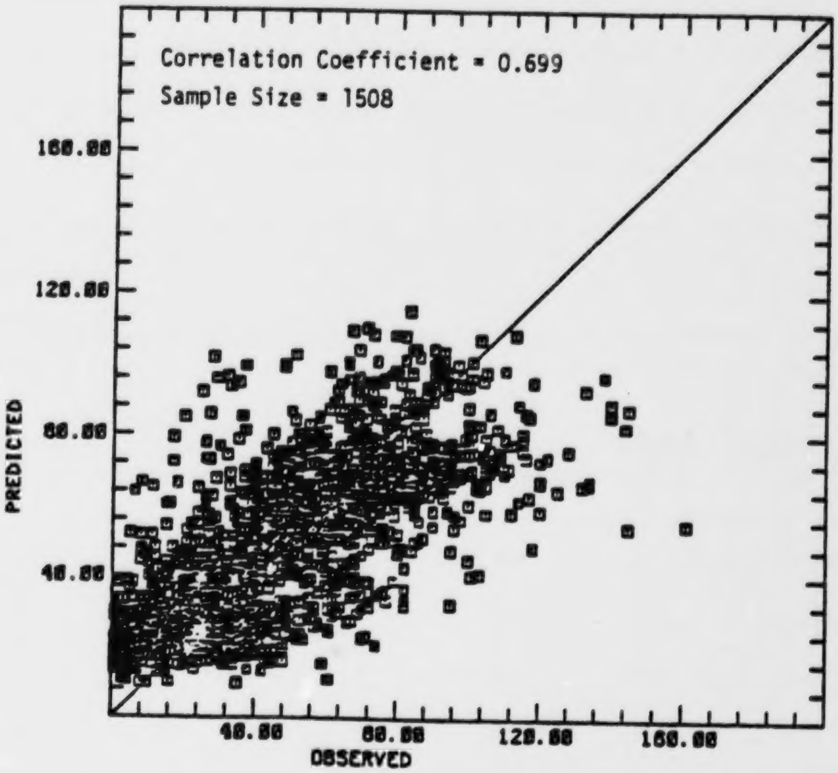


Figure 4. Predicted and observed O_3 concentrations (ppb).

problem also present in the urban photochemical modeling, are apparently attributable to a combination of its short half-life and local contributions.

Figure 5 illustrates the prediction of O_3 distributions over the modeling region. The predicted O_3 patterns, generally corresponding to the prevailing synoptic-scale flows, resemble a southwest-to-northeast O_3 "river" (Figure 5). In two areas, one along the Great Lakes and one near Long Island Sound, the predicted O_3 concentrations exceed 0.15 ppm.

A station-by-station evaluation of the model performance on O_3 predictions is even more impressive. The pertinent statistics are summarized in Table 6. Of the eight stations reporting hourly O_3 concentrations, the correlation coefficients range from 0.67 to 0.88. Concentration histograms for each of the eight stations over the entire 8-day episode are shown in Figures 6 through 13. The station location is indicated in the map by the triangle symbol. The ability of RTM-III to predict the diurnal O_3 patterns at all locations seems to be quite consistent.

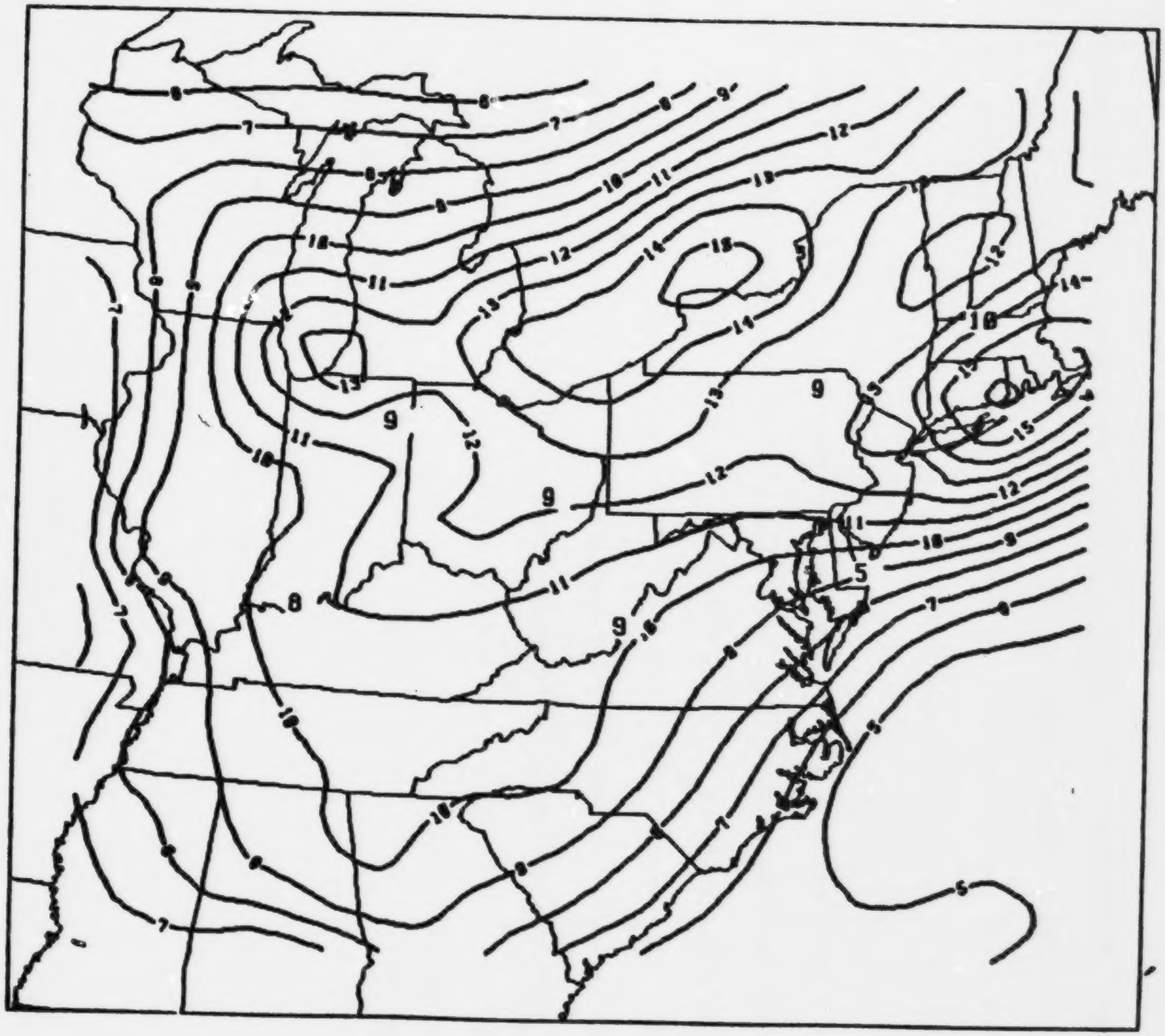

9
9
9
9
5
8
12
12
7
8

Figure 5. Predicted O_3 concentrations in the mixed layer (pphm) on July 20, 1978, at 1400 h.

TABLE 6. MODEL PERFORMANCE STATISTICS FOR HOURLY O_3 BY MONITORING STATIONS[a]

Station Number	Sample Size	Average Observation	Average Bias[b]	Average Absolute Residual	Correlation Coefficient
1	181	55	-1 ± 5	20	0.74
2	171	50	-7 ± 4	20	0.80
3	195	47	-13 ± 4	20	0.72
4	197	51	6 ± 4	18	0.69
5	181	50	7 ± 4	14	0.82
6	198	46	1 ± 4	14	0.74
7	187	46	7 ± 4	15	0.88
8	198	51	8 ± 3	12	0.80

[a] Concentrations in ppb.

[b] At 95% confidence intervals.

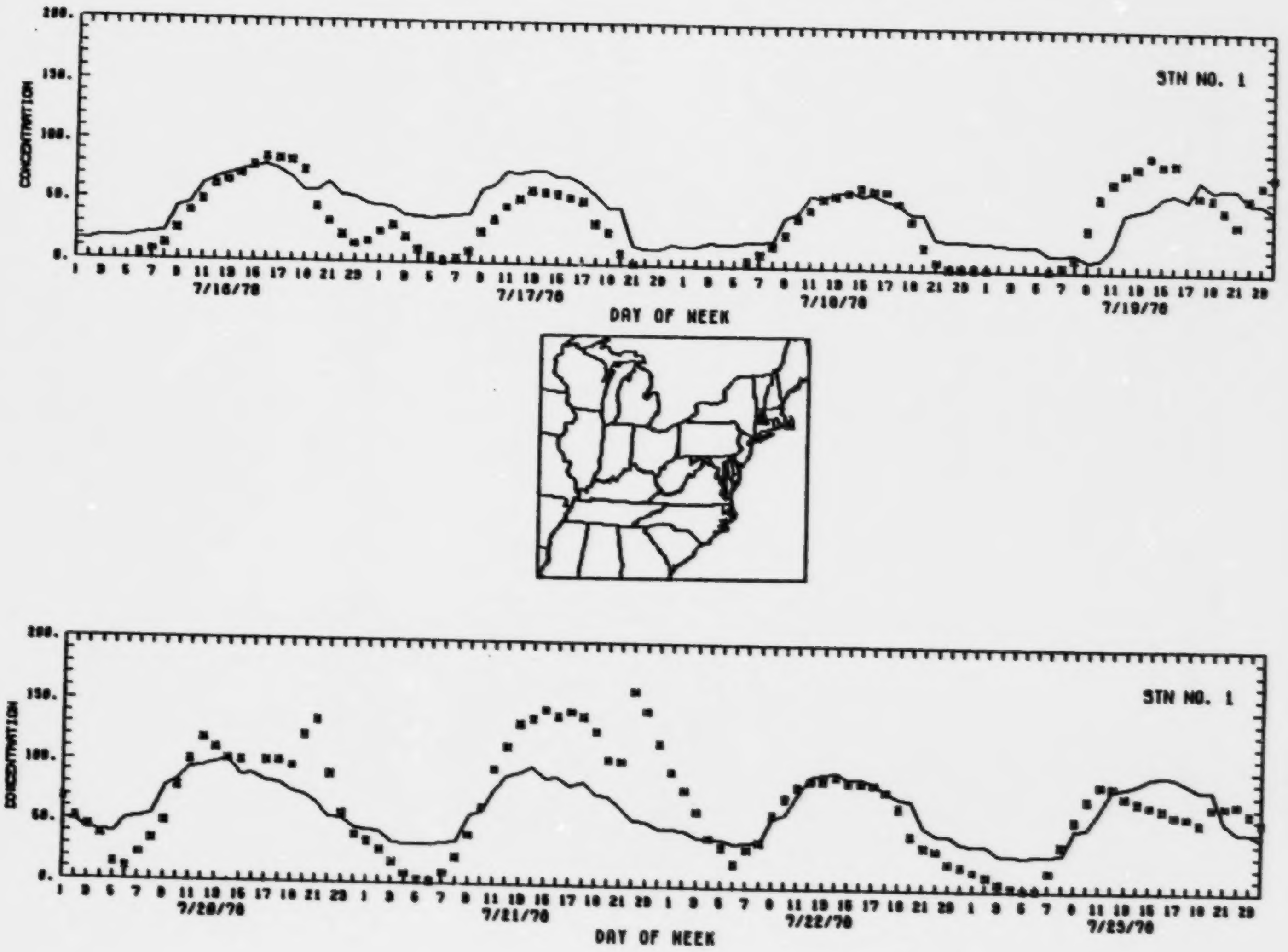


Figure 6. Predicted and measured O_3 concentrations (ppb) at Station 1.

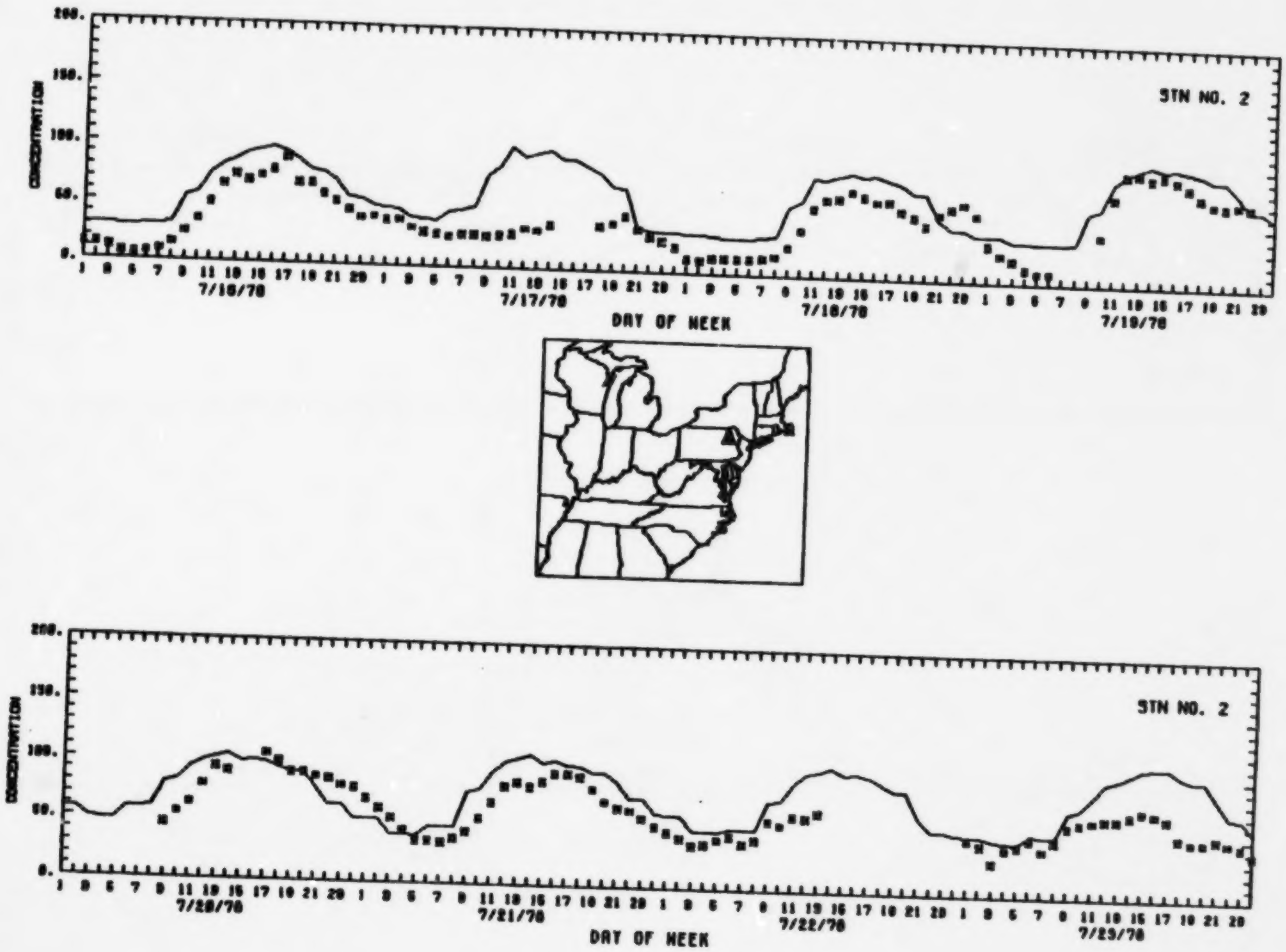


Figure 7. Predicted and measured O_3 concentrations (ppb) at Station 2.

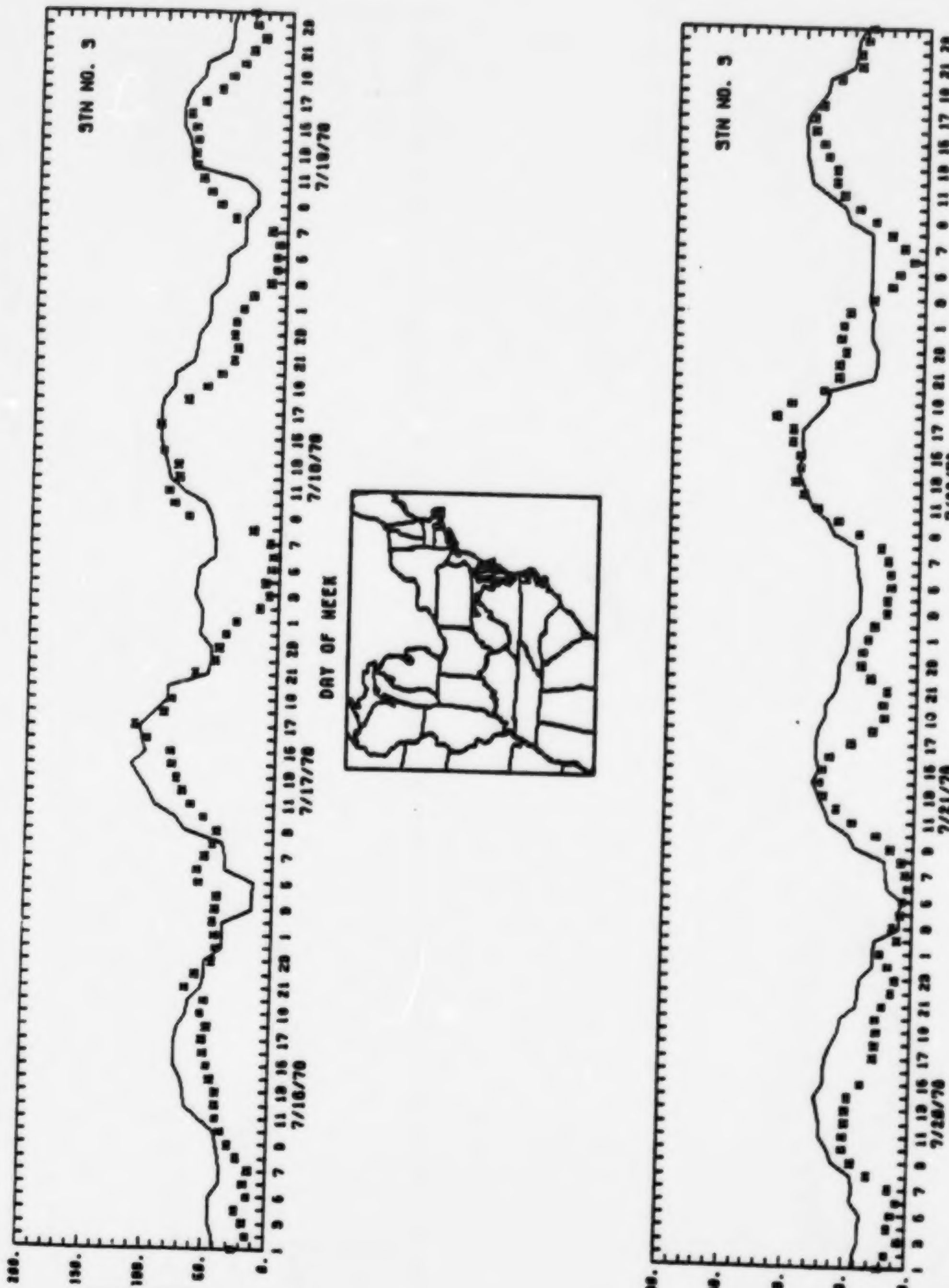


Figure 8. Predicted and measured O_3 concentrations (ppb) at Station 3.

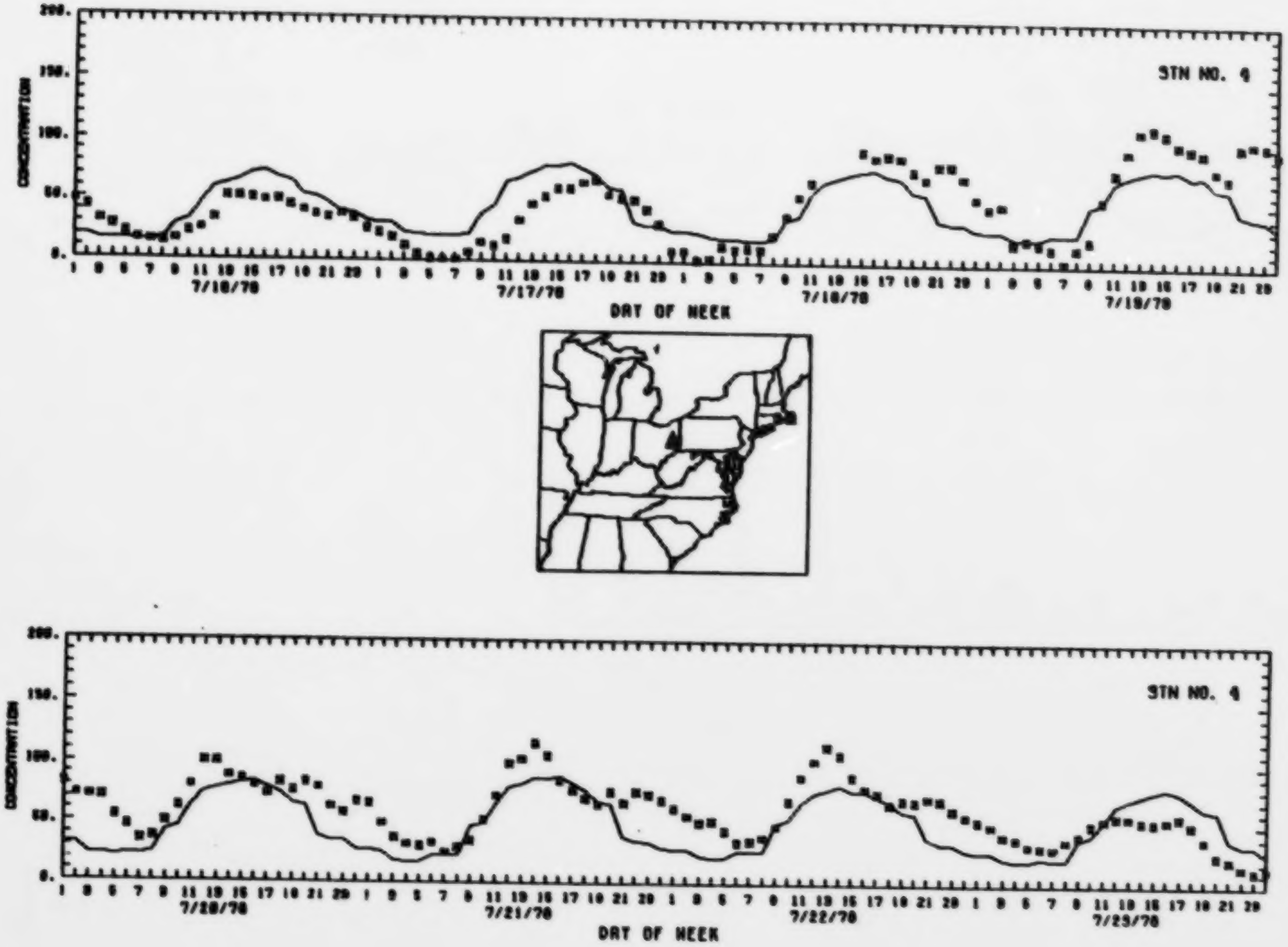


Figure 9. Predicted and measured O_3 concentrations (ppb) at Station 4.

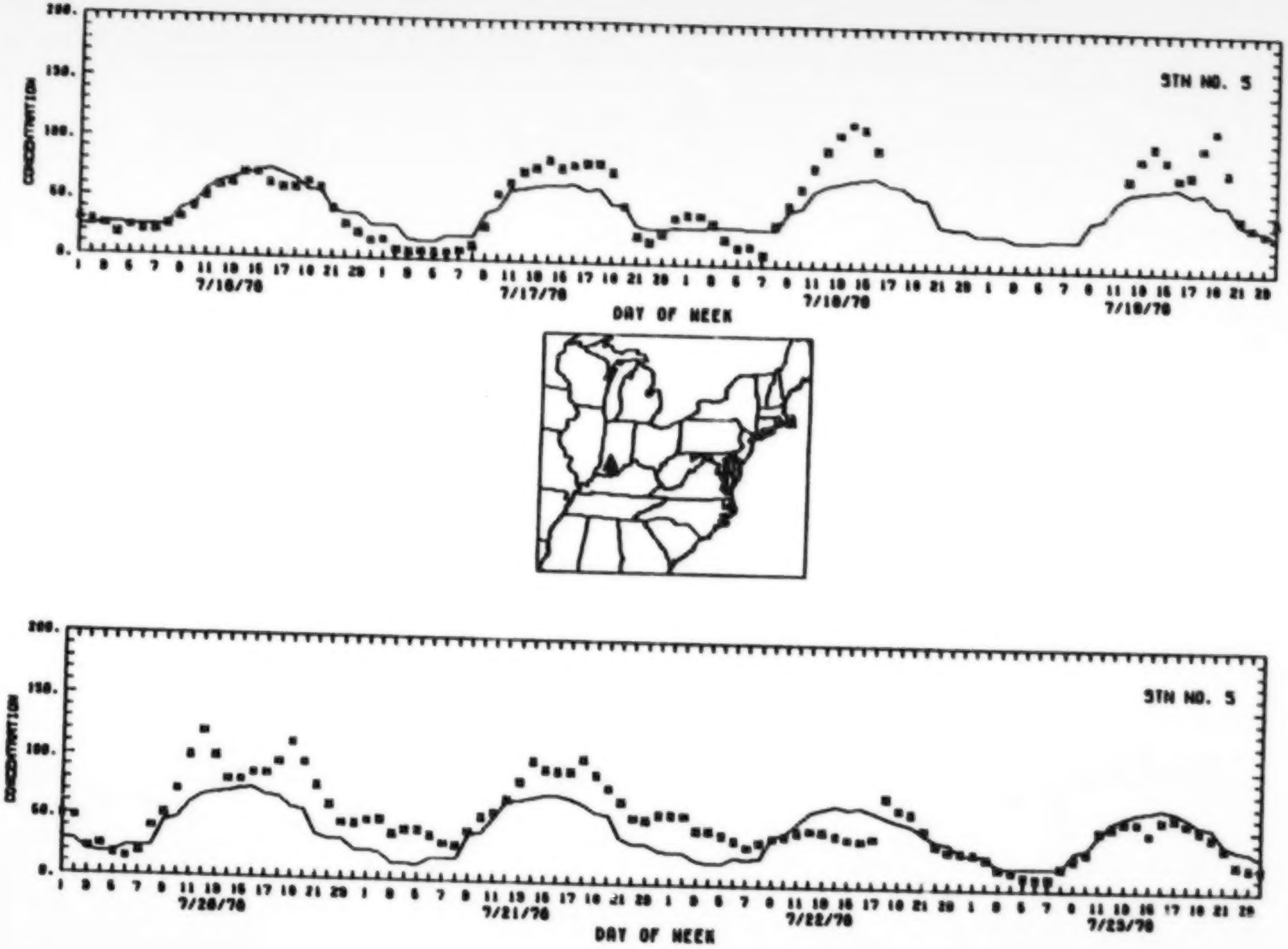


Figure 10. Predicted and measured O_3 concentrations (ppb) at Station 5.

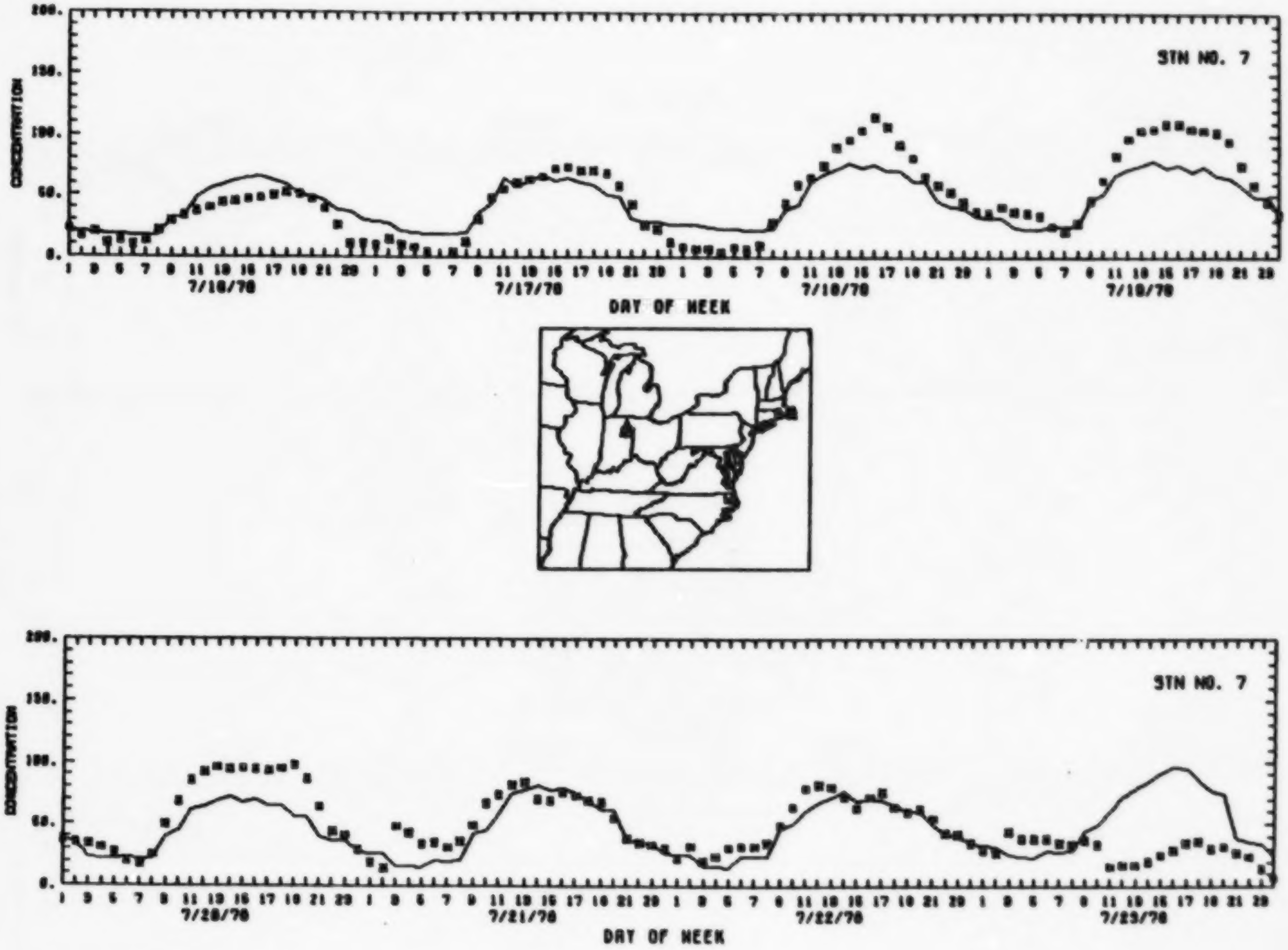


Figure 11. Predicted and measured O_3 concentrations (ppb) at Station 7.

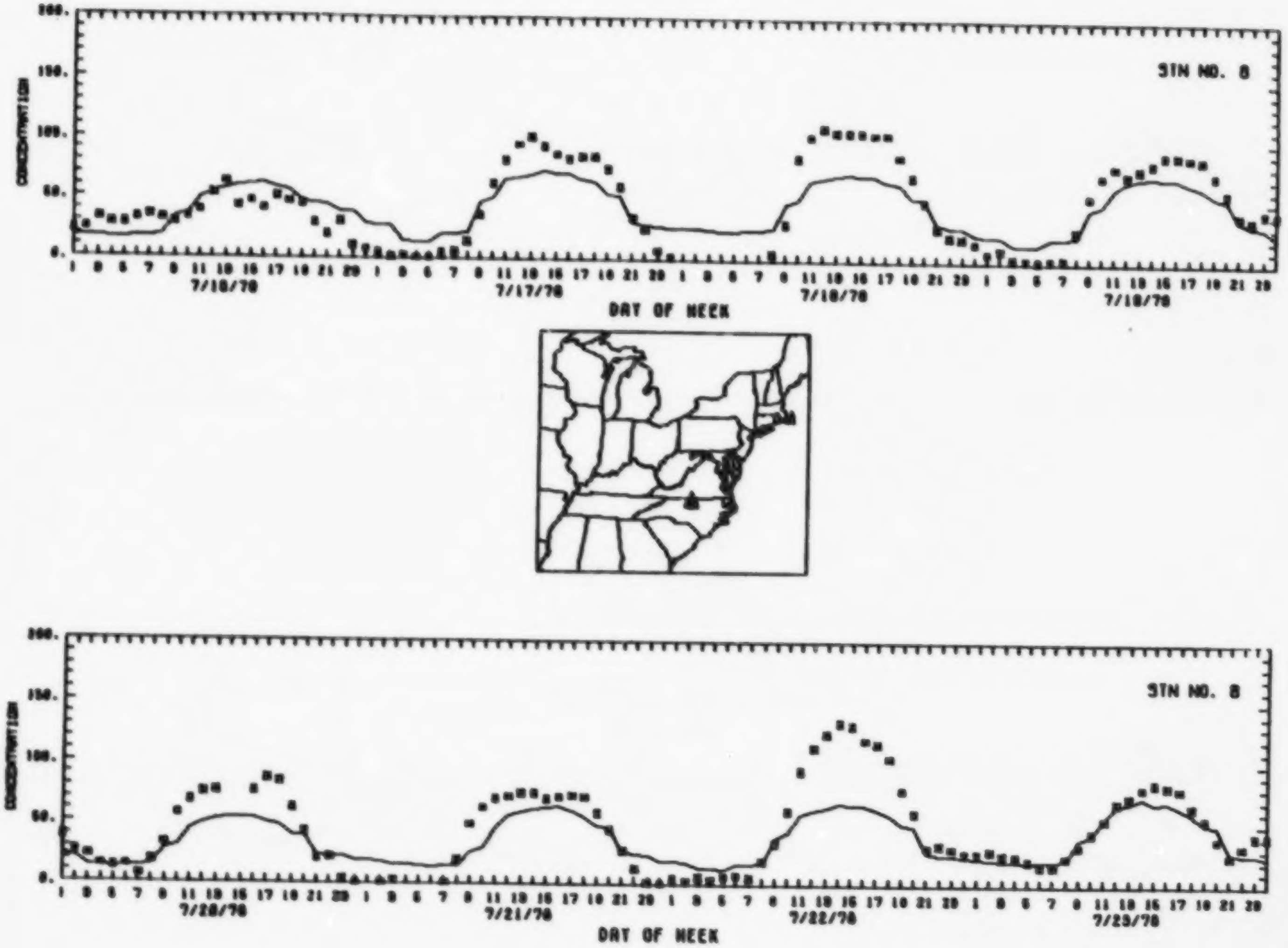


Figure 12. Predicted and measured O_3 concentrations (ppb) at Station 8.

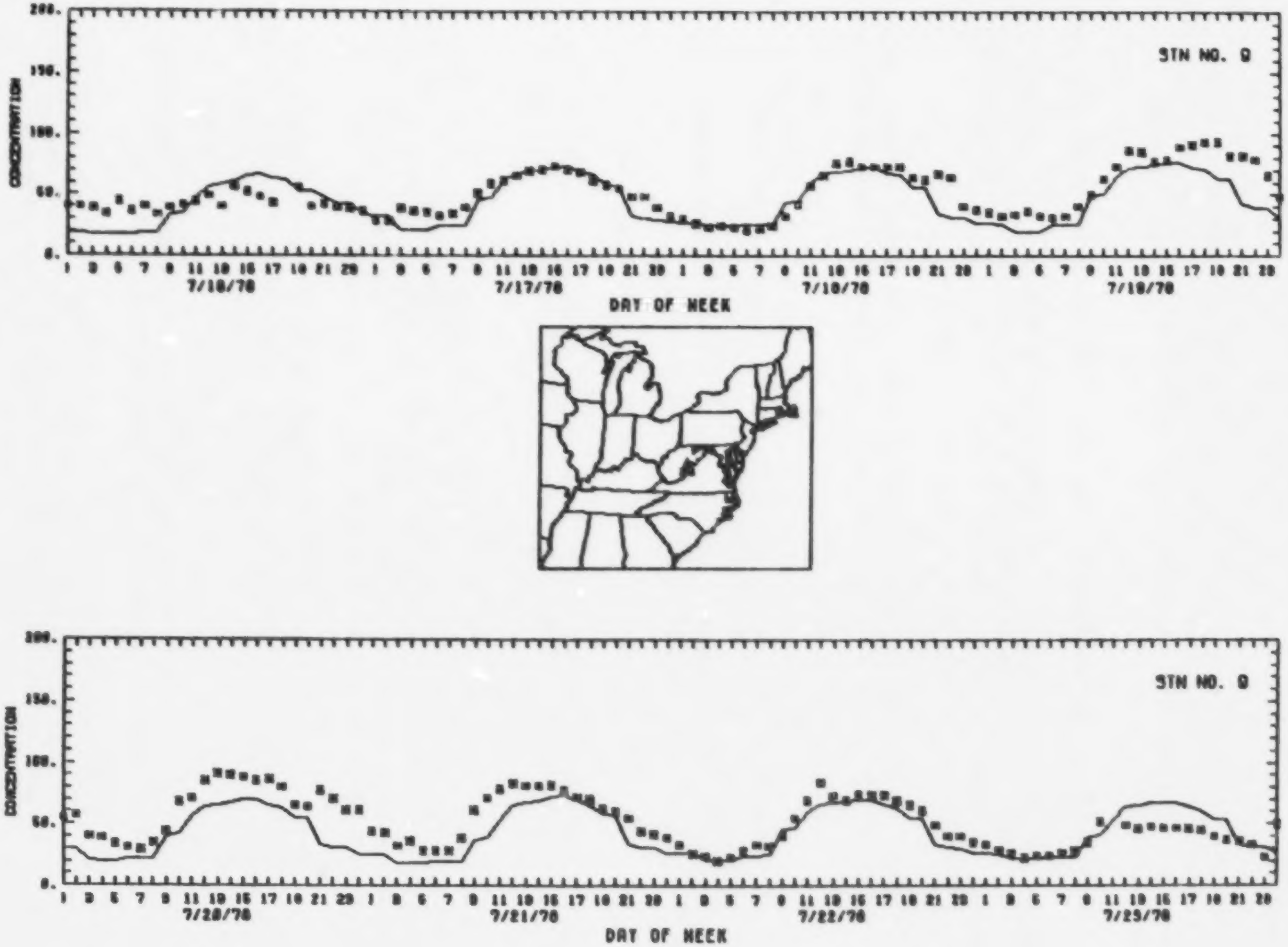


Figure 13. Predicted and measured O_3 concentrations (ppb) at Station 9.

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DISCUSSION

B. Luebkert: Do I understand that you do not use any nighttime chemistry other than deposition or ground scavaging of NO_2 and O_3?

S. Reynolds: That is not correct. We use a very simplified nighttime chemistry. The O_3-olefin reaction still operates. We have largely eliminated most of the olefins from the model and keep those as part of the species anyway, but ethylene is included and the O_3-ethylene reaction does occur at night.

Furthermore, we essentially have a parameterized version of the N_2O_5-and-water reaction operating at night as well.

E. Runca: Are you applying the box model to establish the boundary conditions for the Philadelphia airshed?

S. Reynolds: Correct.

E. Runca: Did you also run the model without these boundary conditions?

S. Reynolds: We did. I do not have those results here, and I am not even sure that they have been released yet. If you are interested in those, I suggest you speak to Dr. Cole.

E. Runca: In what way are the results expected if you compare the simulations?

S. Reynolds: If you compare the simulations to the observations, the simulations without the boundary conditions are obviously much smaller. As Dr. Cole indicated this morning, without the transport, only one of the stations showed an O_3 excess. I can give you what generally happened, and it is as one might expect. The O_3 in this area, which is for the most part upwind of Philadelphia, dropped rather precipitously. Slowly, as one goes farther and farther down into Philadelphia, the O_3 began to increase. The point at which there is relatively little effect of switching off the boundaries is fairly far downwind. I believe the farthest, Dowington Station, was the only one that showed an exceedence about the boundary conditions.

APPENDIX. SAI REGIONAL OXIDANT MODEL (ROM)

II. The SAI Regional Oxidant Model is an Eulerian grid model based on the numerical solution of a 2-1/2 layer multispecies diffusion equation:

1. Variable--time steps vary with wind speed and horizontal resolution (internally controlled by number stability criteria). Output is usually either hourly or 3-h averages. Inputs can be either hourly or 3-h averages.

2. Variable--typical resolution is on the order of 25 km x 25 km.

3. 1,000 km x 1,000 km.

4. Three physical layers--surface layer, generally 10 m to 50 m; mixed layer, variable, 50 m to 1,500 m; inversion layer, variable 50 m to 1,500 m.

5. Top of the inversion layer, generally on the order of 2 km.

6. Inversion layer--geostrophic winds derived from geopotential heights (generally at 850 mbar level).

   Mixed layer--geostrophic wind adjusted for surface effects according to the scheme of Hoxit (1973).

   Surface layer--interpolation scheme of 1/r using surface observations.

7. SHASTA method (Boris and Book, 1973; Book, Boris, and Hain, 1975).

8. Yes. $$\dot{w}_i = -\int_{z_{i-1}}^{z_i} \left(\frac{\partial u_i}{\partial x} + \frac{\partial v_i}{\partial y} \right) dz \qquad i = 1,2,3$$

9. None. Vertical transport and diffusion are treated via interfacial fluxes.

10. $$K_H = \frac{5}{4} (0.28\ \Delta)^2 \left[\left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial y} \right)^2 + \left(\frac{\partial u}{\partial x} - \frac{\partial v}{\partial y} \right)^2 \right]^{1/2}$$

    where Δ is the grid spacing.

11. Assume the pollutant distribution within each physical layer is well mixed.

12. Yes.

    a. Yes
    b. Interpolation of T-sonde measurements
    c. Defines the depth of a homogeneously mixed layer.

13. No.

14. Yes.

15. Yes (see Durran et al., 1979).

    a. Yes.
    b. Yes.

16. Yes.

    a. Yes. Not exercised in the ROM.

17. 2.5 s (CDC 7600)/surface grid point.

18. 64 K small ore, 2.5 Mb disc space (nine species).

    a. CDC 7600.

19. Yes.

    Wojcik, M., T. Myers, J. Killus, O. Serang, and M. K. Liu. 1978. Development and Evaluation of a Mesoscale Photochemical Air Quality Simulation Model. SAI Report No. E178-118, U.S. EPA, 195 pp.

    Liu, M. K., and J. P. Killus. 1981. Development and Evaluation of a Mesoscale Photochemical Air Quality Simulation Model. Proceedings of the 62nd Annual Meeting of the American Association for the Advancement of Science. Pacific Division, Eugene, Oregon.

    Liu, M. K., and P. M. Roth. 1981. The Use of a Regional-Scale Numerical Model in Addressing Certain Key Air Quality Issues Anticipated in the 1980s, In: de Wispelaere, C., ed., Air Pollution Modeling and Its Application, I.

20. Yes.

III. Chemistry

1. Fixed.

    a. O_3, NO or NO_2, CO, PAN, PAR (alkyl carbon atoms), ETH (ethylene), OLE (olefinic bonds), CARB (carbonyl groups, ketones and aldehydes), ARO (reactive aromatics).

b. NO, NO_2, and O_3 are lumped into two species: NO_x (NO + NO_2) and O_x (O_3 + NO_2). NO, NO_2, and NO_3 concentrations are then computed via a steady-state relationship. Radical species O, NO_3, N_2O_5, OH, HO_2, MeO_2, ACO_3, and four intermediate HC-specific peroxy radicals are computed via a steady-state approximation.

2. a. Yes. See above.

   b. Ozone-olefin product chemistry is used; O_3-NO_x chemistry is deleted.

   c. Wind shear and stability stratification are allowed to vary from layer to layer.

3. a. No.

   b. None.

4. Emissions from major point sources are followed by Gaussian puff module (see Liu, M. K., D. A. Stewart, and D. Henderson, 1982, Journal of Applied Meteorology, 21:859-813.

5. No.

6. a. Yes, if input as such.

   b. Cumulus cloud effects can be treated via modifications of photolytic rates.

7. Boundary conditions, initial conditions, source terms.

DEVELOPMENT OF A REGIONAL-SCALE AIR QUALITY MODEL*

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INTRODUCTION

Over the past few years, there has been a marked shift in the emphasis of air pollution problems. Instead of the typically short or episodic problems of a local nature, significant interest has been placed recently on the degradation of air quality or related problems on regional scales caused by the transport of a variety of air pollutants from a large agglomerate of sources over long distances.

During the late 1970s, as part of the plan to promote the use of immense coal reserves in the United States, various potential environmental problems were assessed. Although impacts from primary emissions as a result of coal burning, such as NO_x, SO_2, and particulates, are generally restricted to an area immediately downwind of the sources, problems related to fine particulates and secondary products such as sulfate and nitrate have been raised. These problems have become particularly acute with the increasing use of tall stacks as the control technique for ground-level concentrations, because increasing atmospheric residence times tend to promote chemical reactions.

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*This report has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Even the occurrence of photochemical smog, formerly considered an urban problem, has recently taken on a regional character. For example, in the Northeast Regional Oxidant Study (NEROS), a series of field measurements was conducted to examine the role of pollutant precursors emitted from upwind sources, such as NO_x and reactive HCs, on the high O_3 concentrations observed in rural areas in the Northeast Corridor (Vaughan et al., 1982).

The various air-quality-related problems discussed above have motivated the development of many regional-scale air quality models during the past few years. Several reviews on this subject can be found in the literature (e.g., Eliassen, 1980; Stewart and Liu, 1982). The objective of this paper is to delineate the development and application of a regional-scale air quality model developed by Systems Applications, Inc.

DEVELOPMENT OF A REGIONAL-SCALE AIR QUALITY MODEL

A regional-scale air quality model capable of addressing issues related to long-distance pollutant transport must have the following technical attributes:

- Ability to simulate pollutant concentrations at relatively low levels at locations from 100 to 1,000 km away from the emission sources;
- Ability to simulate physical processes, such as dry deposition, which are only important on large time scales;
- Ability to simulate wet deposition of SO_2, sulfate, and nitrogen-bearing species due to precipitation; and
- Ability to simulate the diurnal formation of secondary pollutants, such as sulfate and O_3 as a result of chemical reactions.

Through a series of research and development contracts, Systems Applications, Inc., has developed a regional-scale air quality model. A brief history of these research and development activities is described in Table 1. This model development, initiated in early 1976, has drawn upon the extensive experience accumulated during the successful development of a photochemical kinetic mechanism (Whitten, Killus, and Hogo, 1980) and an urban airshed model (Reynolds et al., 1973). Over the past 8 yr, the model has undergone evaluation, improvement, and significant expansion. Two different versions of the model now exist:

- A regional transport model for SO_2 and sulfate wet and dry deposition (RTM-II), and
- A regional transport model for photochemical oxidants and their precursors (RTM-III).

The formulations and numerical solutions for these two versions, as described in the next section, are identical.

DESCRIPTION OF MODEL EQUATIONS

The regional transport model described in this paper adopts a puff-on-grid approach to accommodate both point and area sources often encountered in a regional air quality assessment study. The model equations, on an Eulerian grid

TABLE 1. REGIONAL AIR QUALITY MODEL DEVELOPMENT ACTIVITY AT SYSTEMS APPLICATIONS, INC.

Time Period	Activity	Size of Effort (person-years)	Sponsor
1976 - 1977	Development of a regional transport model for SO_2 and sulfate	1	U.S. EPA, Denver, Colorado
1977 - 1970	Development of a regional oxidant model	4	U.S. EPA, Research Triangle Park, North Carolina
1977 - 1980	Development and application of plume and regional visibility model	2-1/2	U.S. EPA, Washington, D.C.
1981 - 1982	Evaluation and improvement of regional transport models	1-1/2	National Park Service, Washington, D.C.
1982 - present	Application of the regional transport model	1-1/4	National Park Service, Washington, Colorado

system, are based on the quasi-three-dimensional, time-dependent atmospheric diffusion equations for multiple chemical species

$$\frac{\partial c_i^j}{\partial t} + u \frac{\partial c_i^j}{\partial_x} + \frac{\partial c_i^j}{\partial y} = \frac{\partial}{\partial_x}\left(K_x \frac{\partial c_i^j}{\partial_x}\right) + \frac{\partial}{\partial_y}\left(K_y \frac{\partial c_i^j}{\partial_y}\right) + E_i^j + F_i^j + R_i^j + Q_i^j - S_i^j \qquad (1)$$

where u and v, K_x and K_y are defined as the horizontal wind velocities and turbulent diffusivities in the x and y directions, respectively. The reaction rate term and pollutant source and sink terms are denoted by R_i^j, Q_i^j, and S_i^j, respectively. E_i^j represents the interfacial transport resulting from entrainment/detrainment and F_i^j the effects of large-scale convergence or divergence, respectively. In this equation, the subscript "i" refers to pollutant species and superscipt "j" denotes the j-th vertical layer.

One of the unique features of this model is the adoption of physical layers rather than predetermined equal- or variable-thickness layers in the vertical direction. As shown in Figure 1, three different physical layers are invoked in the present model. All three layers may vary temporally or spatially. These layers include:

- A mixed layer beneath the inversion layer and above the surface layer that accepts emissions from local sources and carries pollutants trapped below the inversion layer.
- An inversion layer above the mixed layer that serves as a "semipermanent" reservoir for pollutants released from the mixed layer. (The model no longer keeps track of pollutants leaving the top of the inversion layer.)
- A surface layer between the ground surface and the mixed layer that accommodates surface modification of the vertical concentration profiles due to processes such as surface deposition.

The advantage of the present approach is primarily the efficient use of the computational resources. Because no significant concentration variations within the mixed layer are observed at locations distant from the sources, additional layers seem unwarranted. Considerable simplifications can also be realized as coordinate transformations are not necessary when complex topography is encountered.

Relative to horizontal transport by wind, vertical diffusion plays a relatively more minor role than lateral diffusion in determining the fate of air

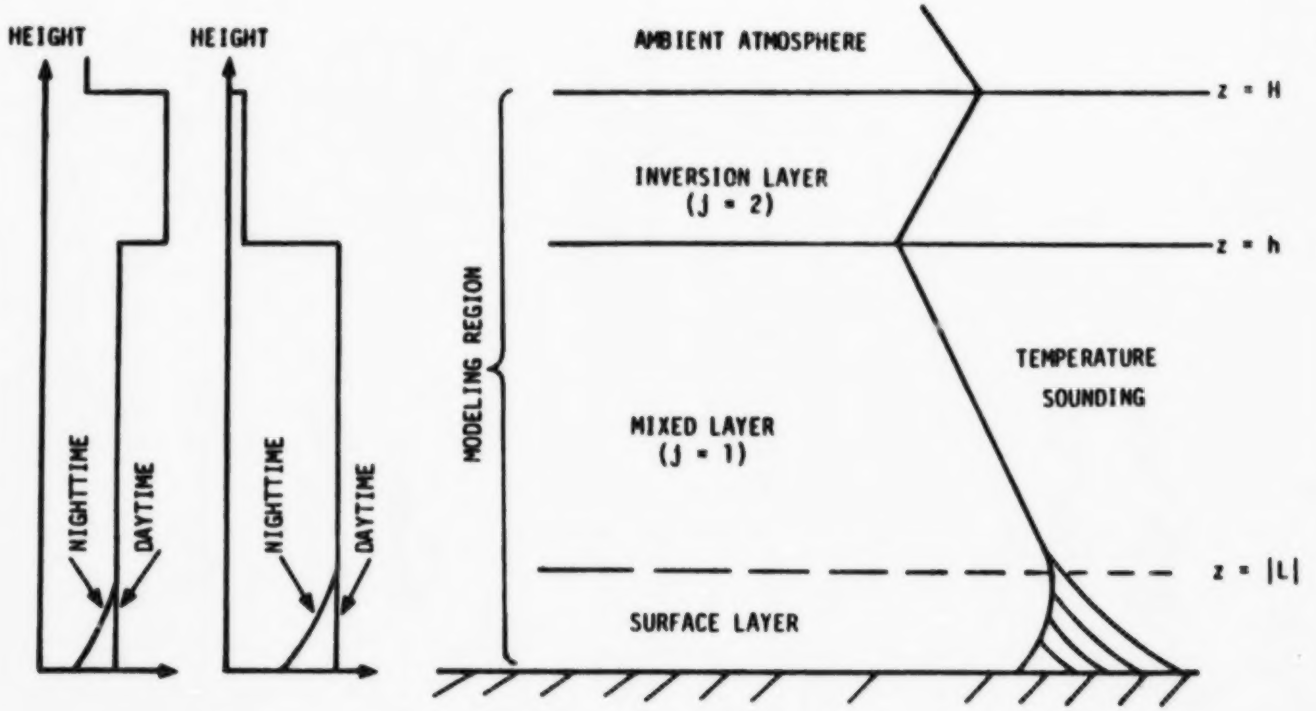

HEIGHT
HEIGHT
NIGHTTIME
DAYTIME
NIGHTTIME
DAYTIME
MODELING REGION
AMBIENT ATMOSPHERE
INVERSION LAYER
(J = 2)
MIXED LAYER
(J = 1)
SURFACE LAYER
TEMPERATURE
SOUNDING
z = H
z = h
z = |L|

Figure 1. Schematics of the vertical structure of the regional transport model.

pollutants on regional scales. This can be seen from a simple dimensional analysis. From the atmospheric diffusion equation, the following two ratios can be formed:

$$\frac{\text{Lateral Diffusion}}{\text{Horizontal Transport}} = \frac{K_H}{U \Delta x}$$

$$\frac{\text{Vertical Diffusion}}{\text{Horizontal Transport}} = \frac{K_V}{U \Delta x} \left(\frac{\Delta x}{\Delta z}\right)^2$$

where U is the characteristic wind speed, and Δx and Δz are the characteristic lengths in the horizontal and vertical directions. According to Heffter (1965) and Randerson (1972), a value of 10^5 m^2/s appears to be the median horizontal diffusivity for the spatial and temporal scales of interest. The vertical eddy diffusivity is a strong function of height and atmospheric stability. For the present analysis, a value of 10^2 m^2/s can be viewed as respresentative (Pasquill, 1974). Thus, using a 10 m/s average wind and Δx = 100 m, the above two ratios become:

$$\frac{\text{Lateral Diffusion}}{\text{Horizontal Transport}} \sim 10^{-1}$$

$$\frac{\text{Vertical Diffusion}}{\text{Horizontal Transport}} \sim 10^2$$

Thus, on regional scales, vertical diffusion can be safely neglected. Vertical exchange of pollutants across the mixed layer and the inversion layer can, however, occur as a result of interfacial transport or entrainment/detrainment.

The mixed layer generally expands or rises during the daytime and collapses or falls in the evening. The resultant entrainment or detrainment of a pollutant across the temporally varying mixed layer is simulated in Equation (1) by the term E_i^j, which can be determined as:

$$
E_i^j = \begin{cases} -\dfrac{\left(c_i^1 - c_i^2\right)}{h}\left(\dfrac{dh}{dt}\right) & \\ \qquad\qquad \text{if } j = 1,\ dh/dt > 0, \text{ or } j = 2,\ dh/dt < 0 & \\ 0 & \\ \qquad\qquad \text{if } j = 1,\ dh/dt \leq 0, \text{ or } j = 2,\ dh/dt > 0 & \\ \dfrac{c_i^2 - c_i^A}{(H - h)}\left(\dfrac{dH}{dt}\right) & \\ \qquad\qquad \text{if } j = 2,\ dH/dt > 0, & \end{cases} \qquad (2)
$$

where h denotes the depth of the mixed layer or the base of the inversion, while H denotes the top of the inversion or the inversion or the height of the entire modeling region and c_i^A is the concentration of pollutant i above the upper boundary of the modeling region.

The fourth term on the right-hand side of Equation (2) represents interfacial pollutant transport into or out of a layer that arises from the large-scale convergence in the spatially varying wind fields. The two-dimensional divergence D is given as $D = du/dx + dv/dy$, and the function $F_i^j(D)$ is defined by:

$$F_i^j = \begin{cases} Dc_i^j & \text{if } D < 0 \\ Dc_i^2 & \text{if } D > 0,\ j = 1 \\ Dc_i^A & \text{if } D > 0,\ j = 2 \end{cases} \tag{3}$$

SIMPLIFICATION OF THE TREATMENT FOR THE SURFACE LAYER

For pollutants originating from either elevated sources or distant ground-level sources, most of the pollutant mass is contained in the mixing layer. The removal processes, as discussed above, consist of diffusion of the pollutants through the surface layer to the ground and absorption or adsorption at the atmosphere-ground interface. As illustrated in Figure 2, through atmospheric stabilities, the diurnal variation of temperature in the surface layer affects the vertical pollutant distribution and, consequently, the rate of surface uptake of pollutants (Hogstrom, 1975). Furthermore, Hill (1971) observed that the adsorption of O_3 by leaves does not vary linearly with concentration at high concentration levels. As a result, a model that can account for these variations must include diabatic atmospheric conditions and nonlinear surface reactions.

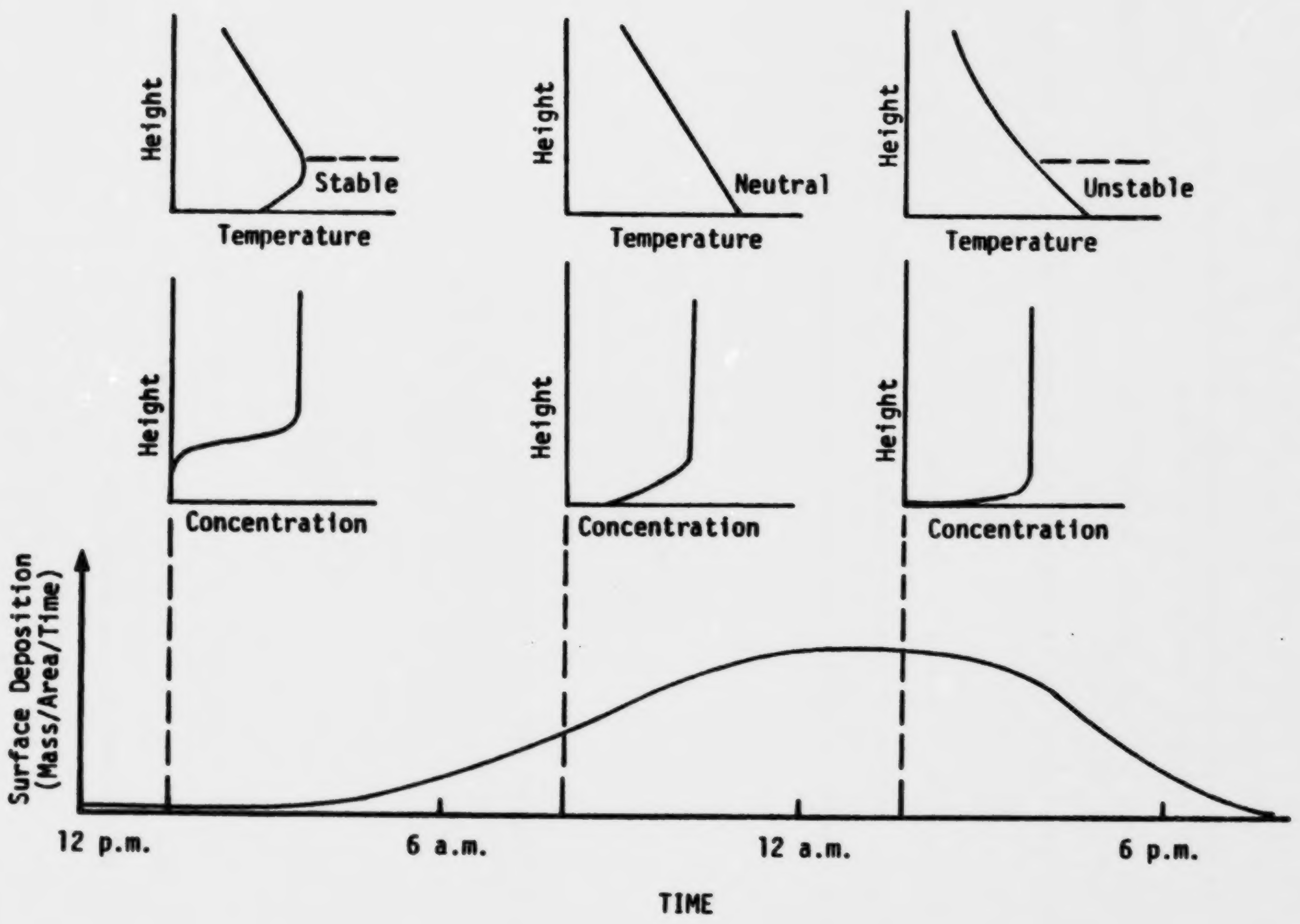


Figure 2. Schematic illustration of diurnal variations in surface deposition.

The model assumes that the transfer of pollutant gases from the atmosphere to a surface is accomplished via three states (Sehmel, Sutter, and Dana, 1973; Galbally, 1974):

- The gases are transported, primarily by turbulent diffusion, to a laminar sublayer just above the surface.
- The gases are transported, primarily by molecular diffusion, through this laminar sublayer.
- The gases interact by adsorption or chemical reaction with the surface.

Thus, as shown in Figure 3, the surface layer is divided into two parts; the turbulent layer and the laminar sublayer. In the turbulent layer, after the atmosphere reaches an equilibrium state, the atmospheric diffusion equation becomes:

$$\frac{\partial}{\partial z}\left[K_v\left(\frac{\partial c}{\partial z}\right)\right] = 0, \qquad (4)$$

with the following boundary conditions:

$$c = c_t \text{ at } z = |L|,$$

$$K_v\left(\frac{\partial c}{\partial z}\right) = f \text{ at } z = z_0,$$

where c_t is the cell-averaged concentration in the mixed layer, f is the pollutant flux across the turbulent layer/laminar sublayer interface, and z_0 is the height of the surface roughness element.

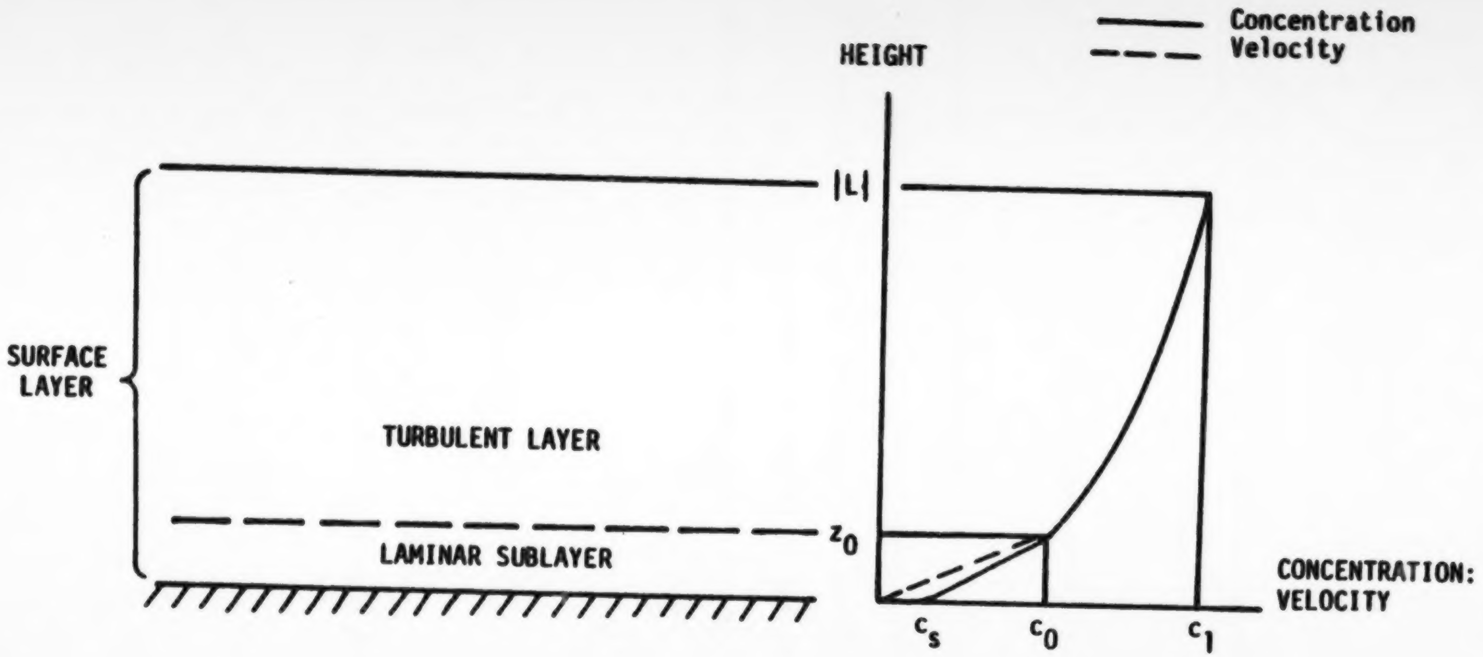

Concentration
Velocity
HEIGHT
|L|
SURFACE LAYER
TURBULENT LAYER
z_0
LAMINAR SUBLAYER
CONCENTRATION: VELOCITY
c_s
c_0
c_1

Figure 3. Schematic illustration of the surface layer.

The vertical diffusivity K_v can be prescribed as:

$$K_v = \frac{ku_* z}{\phi\left(\frac{z}{L}\right)}, \qquad (5)$$

where k = the von Karman constant (0.35),

u_* = the friction velocity,

z = the height, and

L = the Monin-Obukhov length.

This formula is the result of the similarity theory for the constant-flux surface layer. For the neutral case, the ϕ function equals unity. For the stable and unstable cases, the ϕ function is greater and less than 1, respectively. The following empirical expressions for the ϕ function were proposed by Businger et al. (1971) based on the following observational data:

For the stable case ($L > 0$):

$$\phi\left(\frac{z}{L}\right) = 1 + 4.7\left(\frac{z}{L}\right) \qquad (6)$$

For the unstable case ($L < 0$)

$$\phi\left(\frac{z}{L}\right) = \left[1 - 15\left(\frac{z}{L}\right)\right]^{-1/4} \qquad (7)$$

For either the stable or unstable case, the solution of Equation (5) is simply:

$$c = c_1 - f_* \int_{z}^{z_1} \frac{\phi(z')}{k u_* z'} dz' \qquad (8)$$

Across the laminar sublayer, it is assumed that the pollutant flux can be written as:

$$f = \beta u_*(c_0 - c_s) \qquad (9)$$

where c_0 and c_s denote the concentrations at the interface and the surface, respectively; and β, analogous to the Stanton number in heat transfer, is the inverse of a dimensionless resistance for the laminar sublayer.

If it is further assumed that mass and momentum are transferred in an identical manner in the turbulent layer, but differently through the laminar sublayer, then the relationships established by Owen and Thompson (1963) and Thom (1972) discussed above can be used:

$$\beta^{-1} = \alpha \left(\frac{u_* z_0}{\upsilon} \right)^{0.45} \left(\frac{\upsilon}{d} \right)^{0.8} \quad \text{(Owen-Thompson),} \qquad (10)$$

$$\beta^{-1} = \alpha_1 u_*^{1/3} \left[\alpha_2 \left(\frac{\upsilon}{D} \right)^{2/3} - 1 \right] \quad \text{(Thompson)} \qquad (11)$$

Equation (11) is used in the model. To complete the description of the surface-layer model, a boundary condition is required at the surface. Uptake of air pollutants occurs by chemical reaction with, or catalytic decomposition within, either the soil or vegetation, or it can occur by these processes at soil or vegetation surfaces. These processes are generally dependent on the gas concentration at the surface. A general equation for the gas loss per unit of time can be written as (Benson, 1968):

$$f = \gamma \, c_s^{a}, \tag{12}$$

where f is the pollutant flux, γ is a reaction rate constant, and c_s is the concentration of the gas at the soil or vegetation surface. The __________ denotes the reaction order. Eliminating c_0 and c_s from Equations (8), (9), and (10), the following transcendental equation is obtained for f:

$$I \cdot f + \gamma^{-1/a} \cdot f^{1/a} - c_1 = 0, \tag{13}$$

where

$$I \equiv \frac{1}{\beta u_*} + \int_{z_0}^{z_1} \frac{\phi(z)}{k u_* z} \, dz.$$

Although the reaction order is most likely to be 1, closed-form solutions can be found for the cases of a = 1, 2, and 3:

$$f = \begin{cases} \dfrac{c_1}{I + \dfrac{1}{\gamma}} & a = 1 \\[2ex] \dfrac{-\dfrac{1}{\sqrt{\gamma}} + \left(\dfrac{1}{\gamma} + 4Ic_1\right)^{1/2}}{2I} & a = 2 \\[2ex] (A_+ + A_-)^3 & a = 3 \end{cases} \tag{14}$$

where

$$A_{\pm} = \left\{ 3 \quad \frac{C_1}{I} \pm \left[\left(\frac{C_1}{2I} \right)^2 + \frac{1}{27\gamma I^3} \right]^{1/2} \right\}^{1/3}$$

It is interesting to note that these formulas reduce to that of Chamberlain (1966) or Galbally (1974) for the special case of: (1) a first-order surface reaction and (2) a neutrally stratified atmosphere.

CHEMICAL KINETIC CALCULATIONS

The chemical kinetic mechanism used in the regional-scale photochemical air quality model is based on the Carbon-Bond-II mechanism (Whitten, Killus, and Hogo, 1980). This mechanism is expressly carbon conservative, a feature of particular importance in situations involving extended transport and residence times. Hydrocarbon emissions used in the model are divided into five categories, according to the individual bonding structure of carbon atoms within each molecule. The carbon bond categories are: single-bonded carbon (PAR), ethylene (ETH), aromatic rings (ARO), and carbon-fast (OLE) and double-bonded carbonyl (CARB) groups.

The numerical solutions of the rate equations for chemical kinetic mechanisms generally encounter the stiffness problem. Stiff systems are defined as those with widely differing time constants. Classical methods for the solution of differential equations require a time step sufficiently small to avoid instability for the smallest time constant and may impose an enormous computational burden. Although a numerical algorithm based on the method of

Gear (1971) can be used, the introduction of discontinuities into the forcing function of a Gear solution--such as changes in boundary conditions--can cause major inefficiencies in computation. These inefficiencies, together with the additional storage requirements for a high-order predictor-corrector method, make such a scheme unattractive in a grid model.

As an alternative, the invocation of the steady-state aproximation can be used as an important tool for the numerical solution of chemical rate equations. A transformation from differential equations to algebraic solutions allows the reduction of the number of species requiring numerical solutions. The species for which the steady-state approximation is valid are those with the smallest time constants. Thus, an elimination of these species greatly reduces the stiffness of the ordinary differential equations. The complexity of the algebraic solutions increases as more steady-state species that react with each other are included. For example, the implementation of the steady-state approximation in the Carbon-Bond-II mechanism requires the solution of a quintic polynomial for the selected radical species. One problem concerning the use of the steady-state approximation is that its use in effect removes that species from mass balance considerations; i.e., the steady-state expression assumes that $\dot{y} = 0$. If $\int \dot{y} dt$ is very small, then the removal of this species will not affect the overall mass conservation. If, however, $\int \dot{y} dt$ is sufficiently greater than zero, so as to affect the overall mass balance, then the steady-state assumption may produce invalid results.

This limitation has presented a potential problem in the use of the steady-state relationship for simulating major species such as NO, NO_2, and O_3.

Generally, these three species are in dynamic equilibrium, with a time constant much faster than the rest of the nonradical chemical kinetic system. The dynamic equilibrium established is very close to:

$$K_3[O_3][NO] = K_1[NO_2]$$

Unfortunately, mass exchanges involving NO, NO_2, and O_3 are not small, and the conventional form of the steady-state approximation cannot be used. However, the simple steady-state values of NO, NO_2, and O_3 may be modified in terms of a correction factor:

$$\begin{aligned}\Delta_{ss} = -\frac{1}{2}\ [O_3] + [NO] + \frac{K_1}{K_3} \\ + \frac{1}{2}\left\{\left([O_3] + [NO] + \frac{K_1}{K_3}\right)^2\right. \\ \left. - 4\left([NO][O_3] - \frac{K_1}{K_3}\ [NO_2]\right)\right\}^{1/2}, \qquad (15)\end{aligned}$$

where

$$[NO_2]_{ss} = [NO_2] - \Delta_{ss}$$

$$[NO]_{ss} = [NO] + \Delta_{ss}$$

$$[O_3]_{ss} = [O_3] + \Delta_{ss}$$

For state variables, such as NO_x (=NO + NO_2) and unpaired oxygen atoms O_x (=O_3 + NO_2), the correction-factor equation becomes:

$$\Delta_{ss} = -\frac{1}{2}\left([O_x] + [NO_x] + \frac{K_1}{K_3}\right)$$
$$+ \frac{1}{2}\left\{\left([O_x] + [NO_x] + \frac{K_1}{K_3}\right)^2 - 4\,[NO_x]\,[O_x]\right\}^{1/2},$$

where

$$[NO_2]_{ss} = -\Delta_{ss}$$
$$[NO]_{ss} = NO_x + \Delta_{ss}$$
$$[O_3]_{ss} = O_x + \Delta_{ss}$$

Mass conservation is thus maintained for the two redefined species, NO_x and O_x. The steady-state calculation does not affect the quantity of these two species; it merely apportions them into the three molecular species O_3, NO, and NO_2. In this scheme, NO-to-NO_2 conversions become the source of unpaired oxygen atoms.

This scheme has been tested against the Gear solution for a variety of cases. As might be expected, the approximation tends to break down when peroxyl radical concentrations are large relative to O_3 (i.e., when there are very high HC concentrations or very low NO_x concentrations). The former condition is not likely to be encountered, given the resolution of a regional-scale model. Under the latter condition, photochemical O_3 production is very slow relative to the background. Thus, the approximation appears to be reasonable for regional-scale application. The incorporation of a similar scheme in the Regional Oxidant Model (RTM-III) for simulating aerosol formation is currently being contemplated. A description of the proposed aerosol module is described in the appendix.

APPLICATIONS OF THE REGIONAL TRANSPORT MODEL

In this section, we will summarize two typical applications of the regional transport model described in the previous sections. A preliminary version of the regional photochemical air quality model (RTM-III) was applied to the Northeast Regional Oxidant Study (NEROS) areas. The SO_2/sulfate version of the regional transport model (RTM-II) was applied to a sulfate episode in July 1978, reported by the Sulfate Regional Experiment (SURE) in the Northeast United States.

Application of the NEROS Areas

As a result of the long-range transport of O_3 and its precursors, elevated oxidant concentrations have been observed in many rural areas in the northeastern part of the United States. Measurements collected at remote locations, such as the White Mountains, indicate O_3 concentrations significantly higher than the background levels (Lonneman, 1977). Observations drawn from the aircraft data reported by Simple et al. (1977) show that O_3 concentrations at an elevation of 2 km are consistently between 40 and 60 ppb, regardless of the concentrations below this level. Exceptions occur when the base of the synoptic subsidence inversion is above 2 km. On these occasions, O_3 values at 2 km can reach 80 ppb. Both observations seem to imply that the high O_3 levels were caused by the transport of pollutant precursors from distant upwind sources. A preliminary version of the regional photochemical air quality model (RTM-III) was applied to this area (Wojcik et al., 1978). The model region covered an area approximately 800 km x 800 km, with a spatial resolution of 20 km and a temporal resolution of 3 h. Data collected during the 1975 Northeast Oxidant Transport Study were used for testing and validating the model.

For the modeling region, gridded area emission rates for particulates, SO_2, NO_x, HCs, and CO were derived from the National Emissions Data System (NEDS) on a 40-km x 40-km grid with a 20-km resolution. To conform to the modeling grid, these emissions were interpolated to the grid by proportionally allocating the emissions from each cell. In addition to area sources, emission rates for point sources exceeding 10,000 tons/yr were also included. Although most emissions were yearly averages, some data were available for seasonal variations and the number of hours of operation per day. The hours of operation were assumed to be symmetrically distributed around noon and to have equal hourly emission rates. For point sources having no data for hours of operation, the emission rates were assumed to be constant, with the exception of power plants.

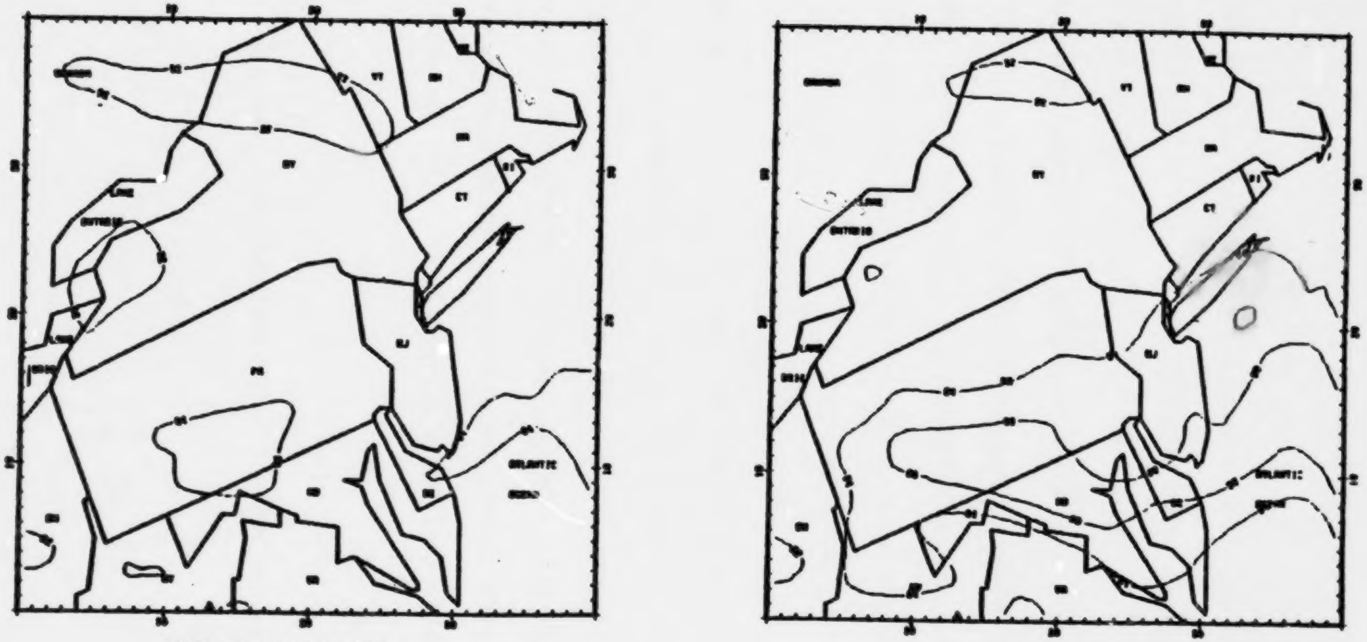
The model was exercised for 72 h between 0100 EDT on July 29, 1975, and 0100 EDT on July 31, 1975. The model predictions of primary pollutants such as NO and SO_2 are easily discernible and generally show average concentrations in the mixed layer that increase near major sources. These areas include the industrial areas extending from New York City to Southern New Jersey, Northern Delaware, and Southern Pennsylvania (Philadelphia). Also apparent are the

Washington, DC/Baltimore urban complex and several major point sources in New York, Pennsylvania, and West Virginia.

An analysis of the prediced O_3 patterns is interesting. As shown in Figure 4, O_3 concentrations before 0600 EDT on July 30, 1975, are generally less than 60 ppb. The level increases to a peak of 120 ppb during late afternoon, with the high O_3 concentrations areas extending from south Pennsylvania to the Atlantic Ocean. After sunset, the predicted O_3 concentrations begin to decrease to a level generally lower than 80 ppb. With a similar diurnal pattern, the predicted O_3 concentrations reach a maximum of 140 ppb near New Jersey between 1800 and 1900 EDT on July 31, 1975.

For comparison, observed surface O_3 concentrations from 1300 to 1500 EDT on both days are displayed in Figure 5. On July 30, high O_3 concentrations ranging from 100 ppb to 150 ppb were reported along a corridor from Wilmington, New Jersey, to New York City. On July 31, the observed O_3 concentrations generally increased, with high O_3 areas extending further northeast. Thus, a good qualitative comparison of the model predictions with the observation is achieved.

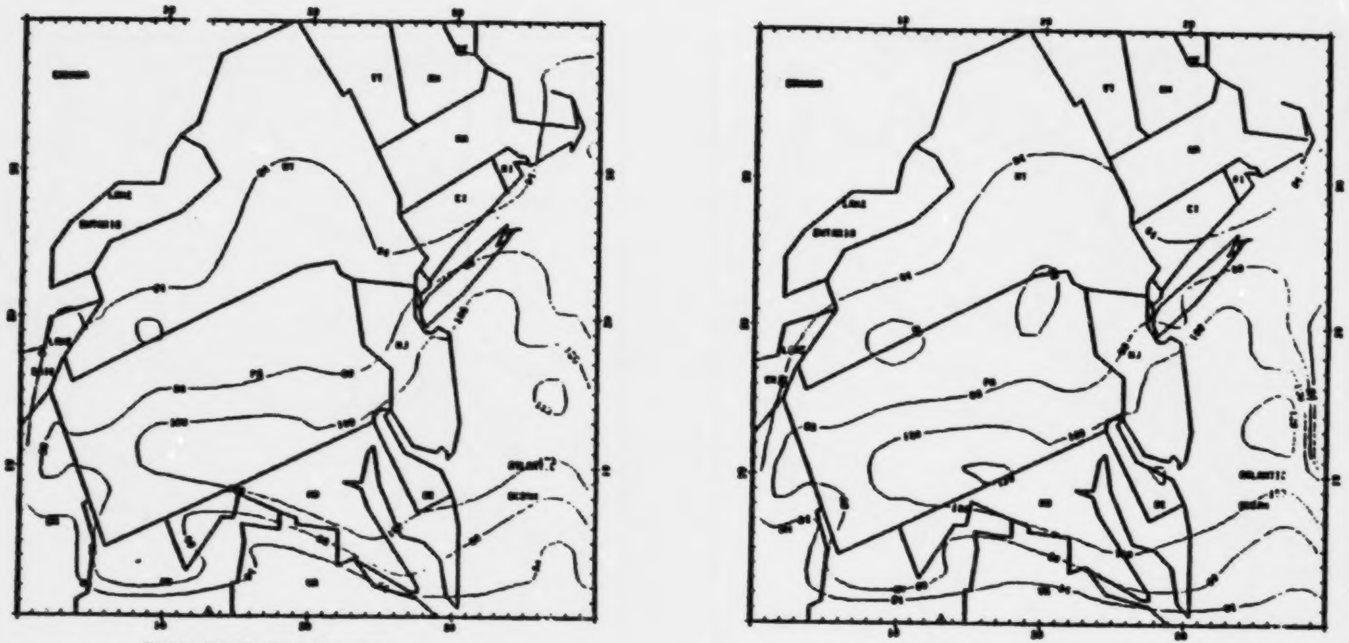
Further application of RTM-III using the SURE data base will be discussed by Killus et al. (1983).


VT
NY
CT
RI
NJ
PA
LAKE
ONTARIO
ATLANTIC
OCEAN

0600-700 EDT 750730

1000-1100 EDT 750730

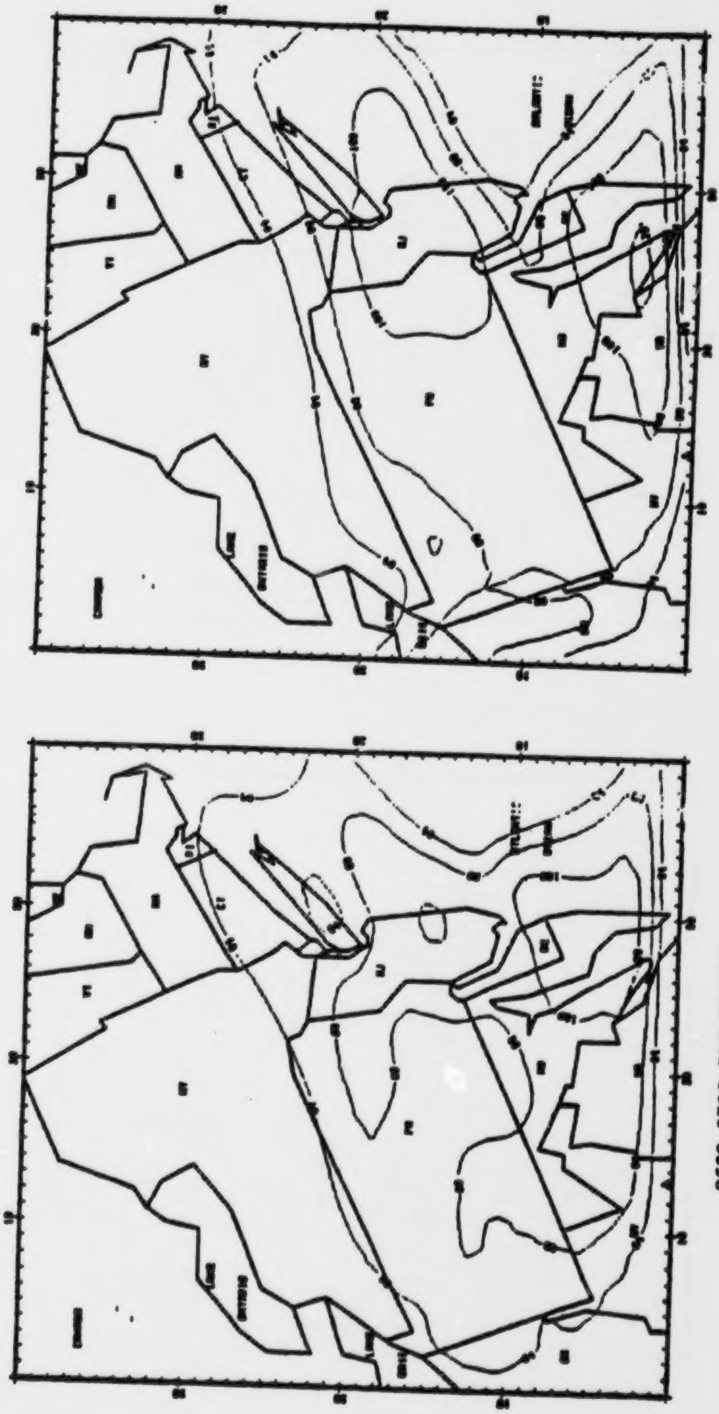
Figure 4. Predicted O_3 distributions in the Northeast United States (concentrations in ppb).



1400-1500 EDT 750730

1800-1900 EDT 750730

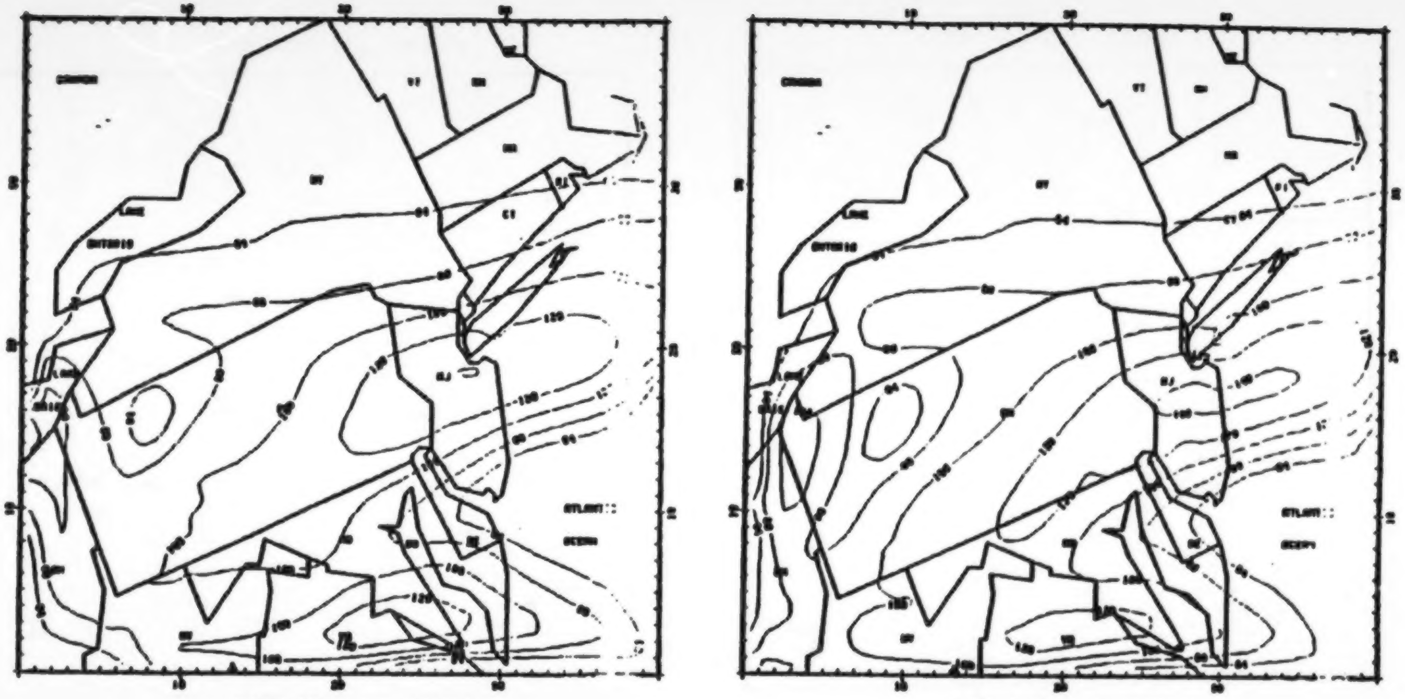
Figure 4. (continued).



0600-0700 EDT 750731

1000-1100 EDT 750731

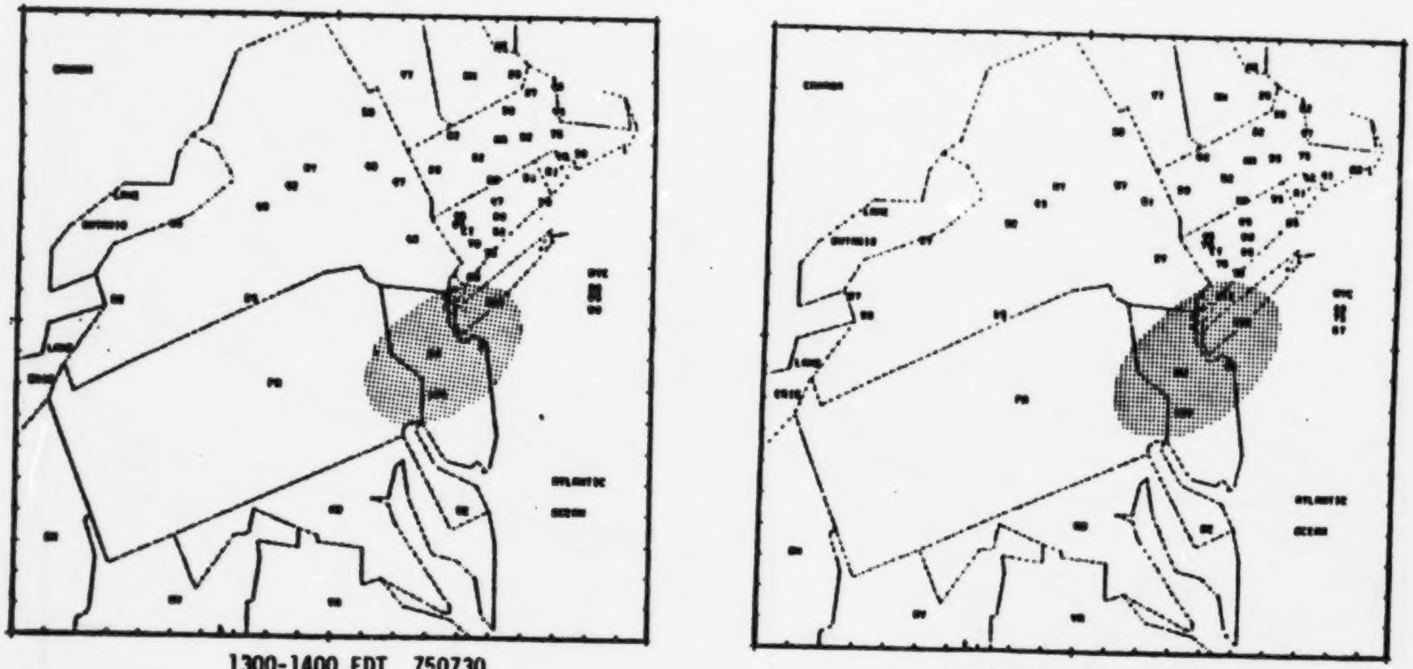
Figure 4. (continued).



1400-1500 EDT 750731

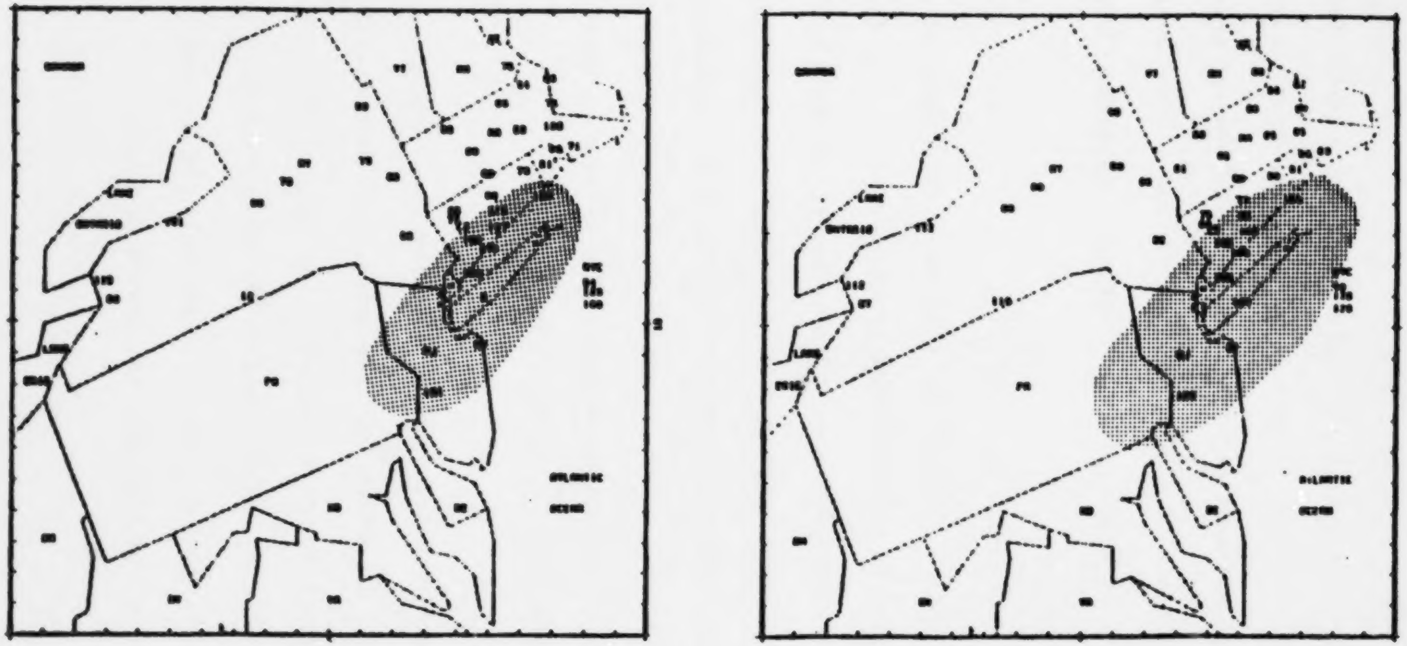
1800-1900 EDT 750731

Figure 4. (continued).


1300-1400 EDT 750730

1400-1500 EDT 750730

Figure 5. Observed surface O_3 concentrations in the Northeast United States (concentrations in ppb).



1300-1400 EDT 750731

1400-1500 EDT 750731

Figure 5. (continued).

Application to a Sulfate Episode

The SO_2/sulfate version of the Regional Transport Model (RTM-II) was applied to a sulfate episode in July 1978 using data from the EPRI Sulfate Regional Experiment (Stewart et al., 1983). The purpose of this study was to evaluate the performance of the model using SO_2 and sulfate data collected at 54 monitoring stations geographically distributed throughout the modeling region.

The modeling region selected for this study was almost identical to the EPRI/SURE grid, which is characterized by 80-km mesh squares defined over the eastern third of the United States. Dimensions of the grid subset used for this study were 2,080 km in the east-west direction by 1,840 km in the north-south direction. The grid resolution selected for the model simulations was 40 km x 40 km. Wind velocities and mixing depths were derived from the National Weather Service (NWS) radiosonde network. The emissions inventory used in this study was prepared as part of the EPRI/SURE program.

RTM-II was exercised for a period spanning eight consecutive days (July 16-23, 1978). To ensure that the sulfur mass budget within the modeling domain reached a quasi-steady state before evaluation, the model was exercised for 48 h prior to July 16, 1978. Temporally varying boundary conditions during the simulation period were estimated by extrapolating average 3-h concentrations from a few monitoring stations near the upwind boundaries.

Figures 6 through 10 are a series of time-history plots for the model predictions and corresponding observations at each of the SURE Class I and Class II stations throughout the modeling region. The predicted 24-h average concentrations are displayed as dotted lines, and the observed 24-h average concentrations are indicated by square symbols. Successive figures include stations grouped geographically from the western to eastern (generally downwind) portion of the modeling region.

An examination of these plots suggests that the model's ability to predict the sulfate trend is quite good. The ability of the model to simulate the SO_2 trend appears to be less favorable, a fact that may be attributable to the influences of local emission sources. A common characteristic of both SO_2 and sulfate predictions is that the temporal variations are smoother than their observed counterparts. This is probably caused by several factors, including the spatial resolution of the model, which tends to smooth out localized concentration peaks.

The relatively smooth sulfate predictions produce a less pronounced jump in concentrations, most noticeable at Station 29 (Figure 7) and Stations 30 and 14 (Figure 8). Similarly, the model tended to underpredict the sulfate peak in Illinois on July 18, 1978 (Stations 26, 27, and 38 in Figure 6). Further analysis indicates that the high sulfate region in Illinois (>30 $\mu g/m^3$) was associated with a weak anticyclonic flow centered in western Kentucky and Tennessee. An examination of the back-trajectories shows emissions from the St. Louis area and the Ohio River Valley were advected over central Illinois at

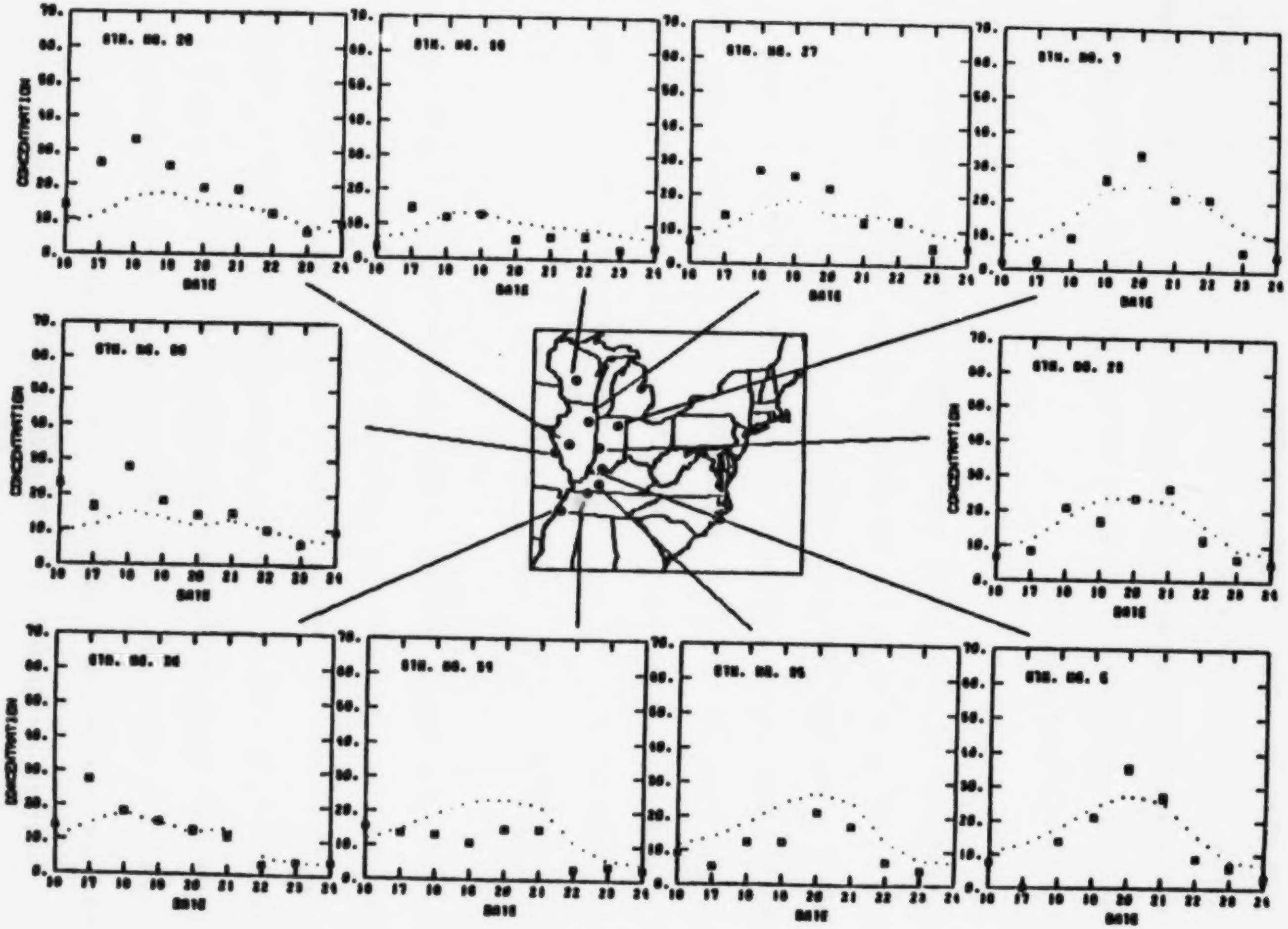


Figure 6a. Time-history plot of daily sulfate and SO_2 concentrations for the 10 westernmost monitoring stations: (a) Sulfate concentrations, (b) SO_2 concentrations. Predicted = ..., and measured = ■.

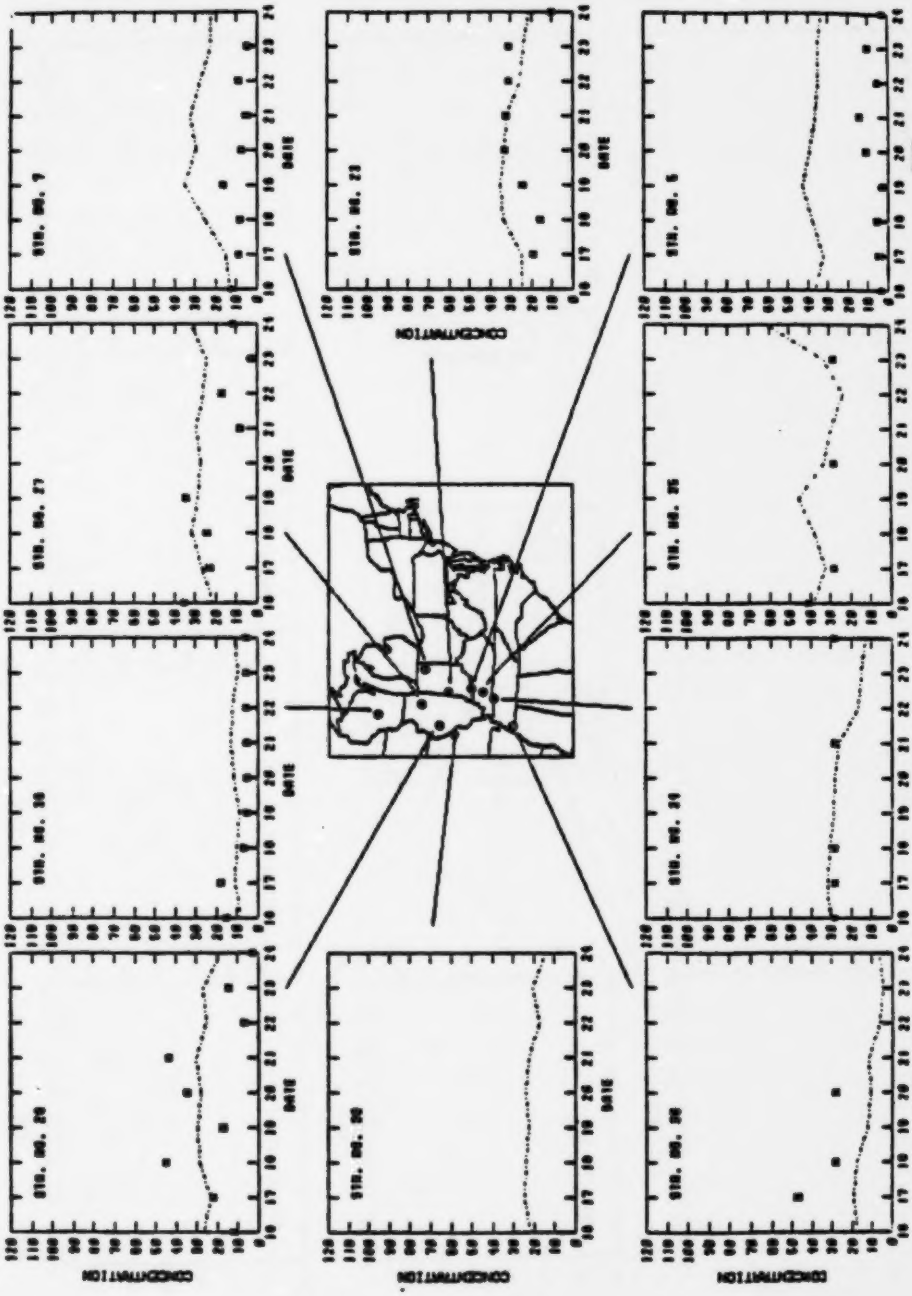


Figure 6b. (continued).

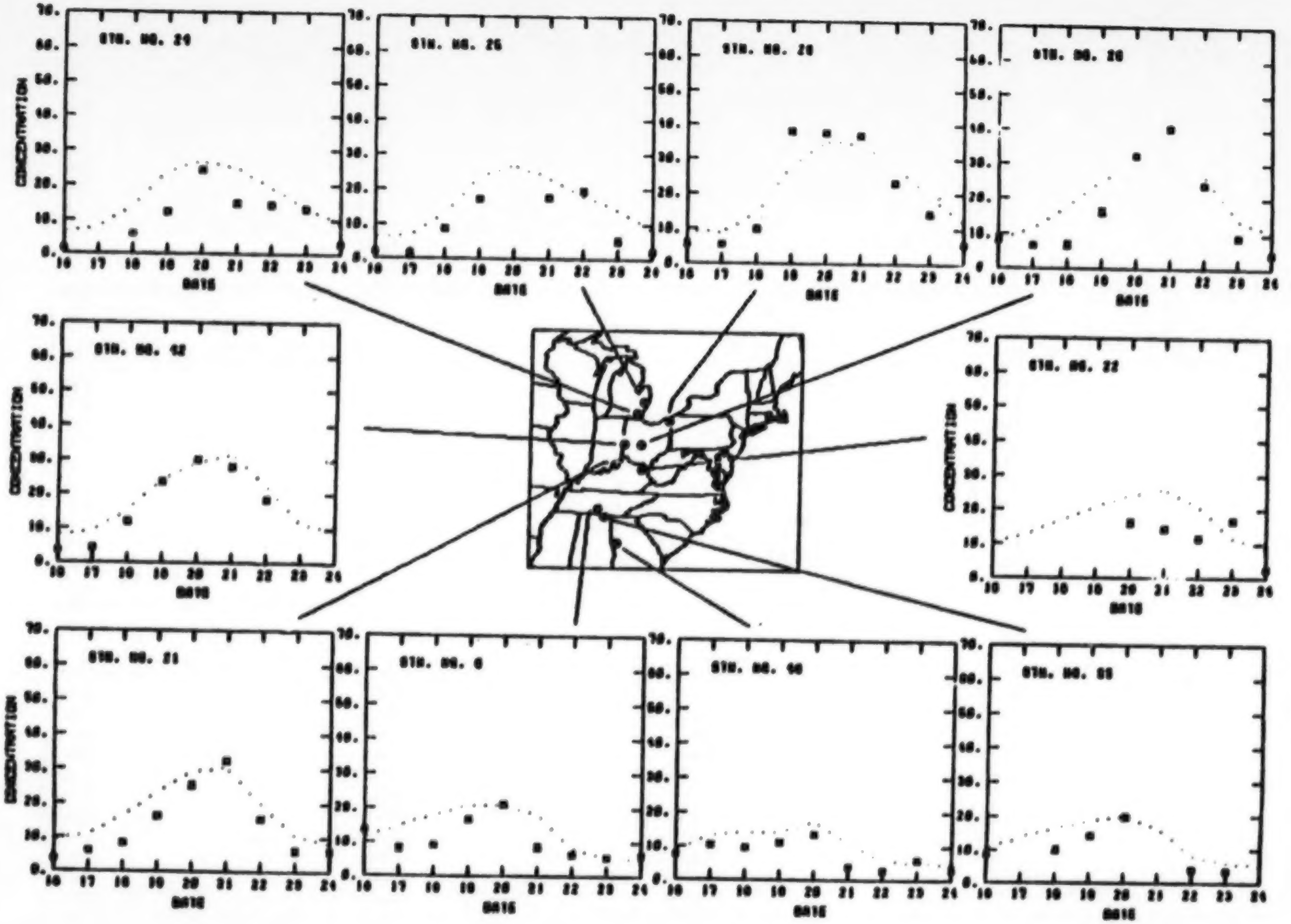


Figure 7a. Time-history plots of daily sulfate and SO_2 concentrations for the 10 westernmost monitoring stations: (a) Sulfate concentrations, (b) SO_2 concentrations. Predicted = ..., and measured = ■.

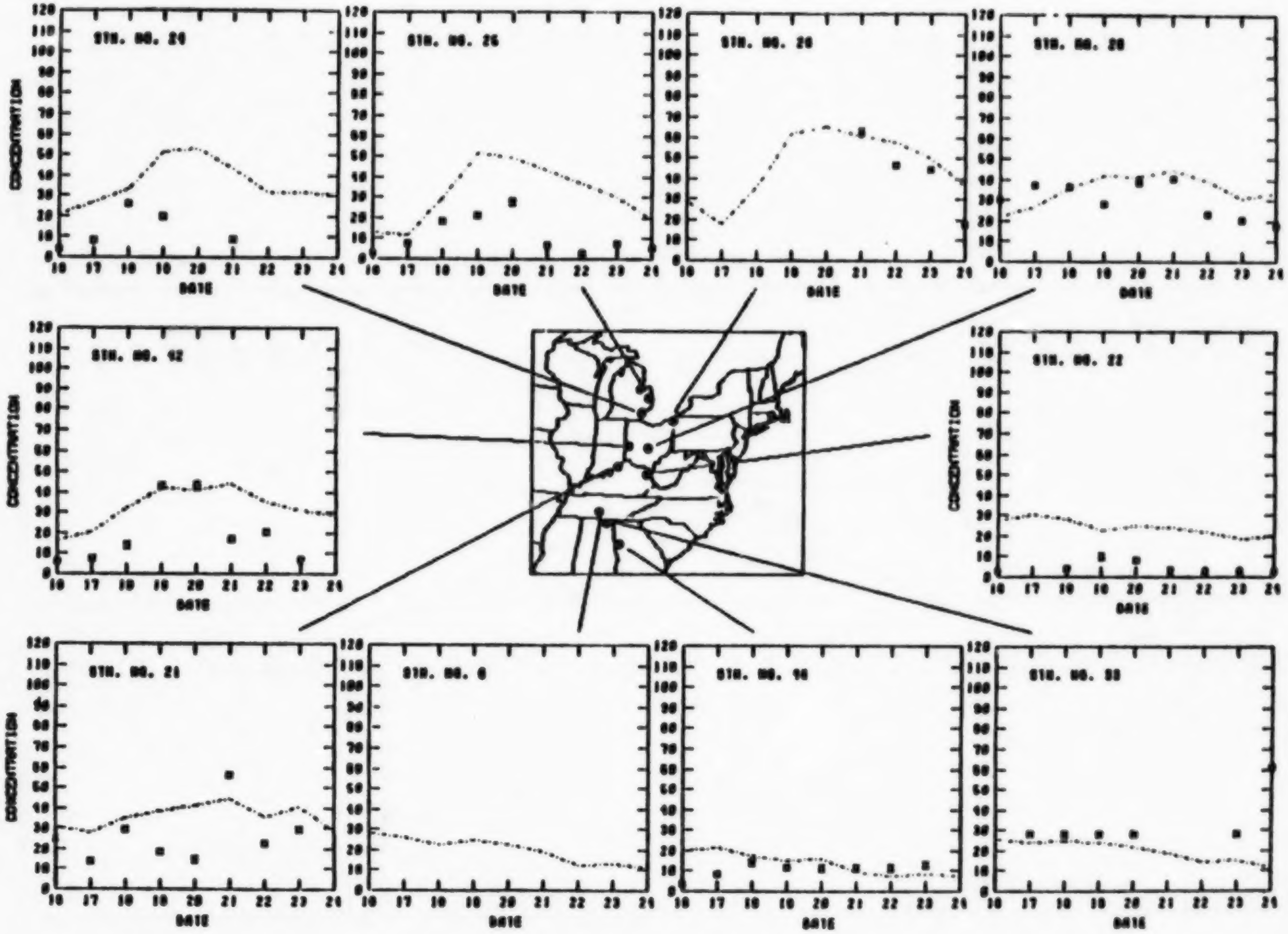


Figure 7b. (continued).

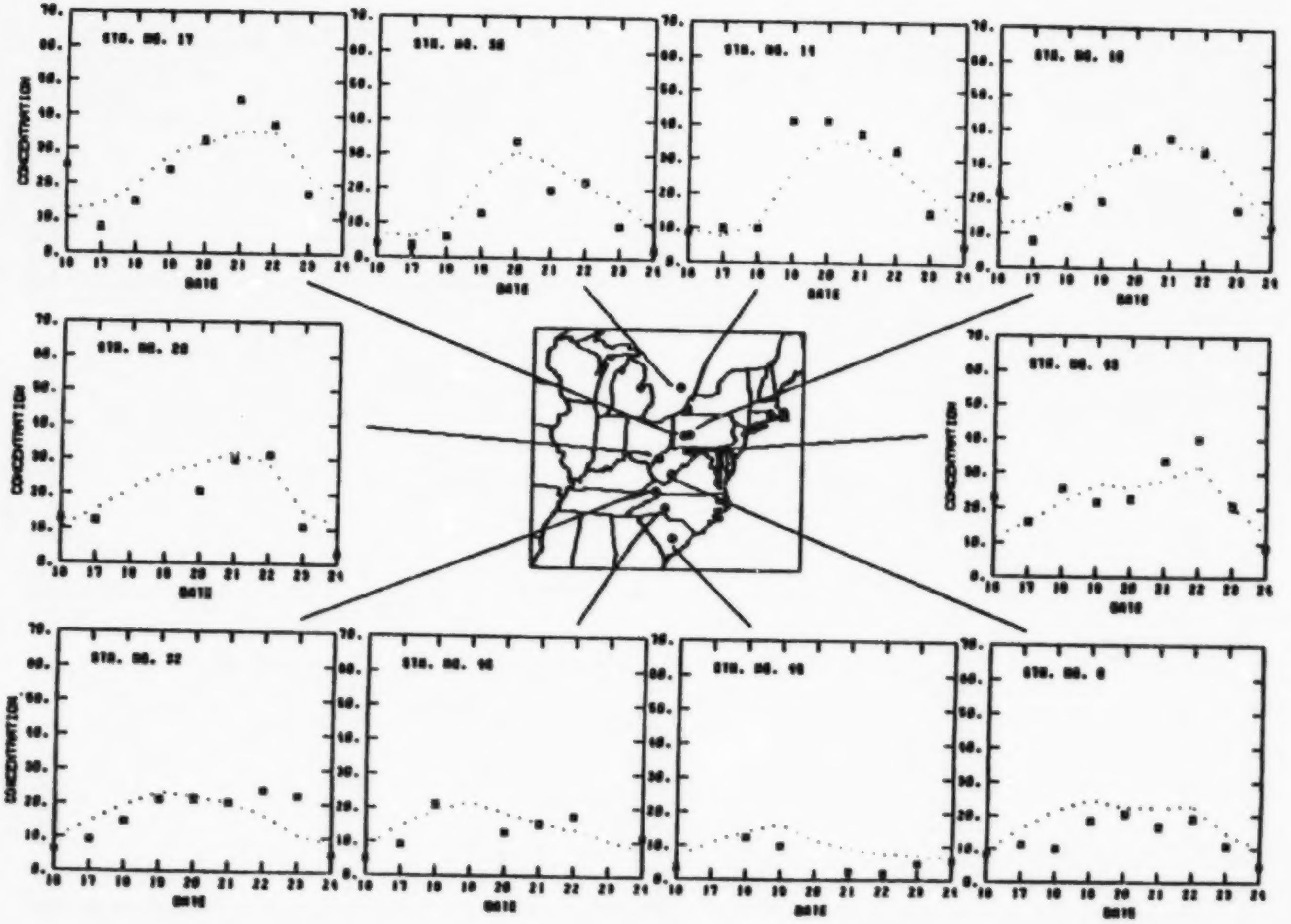


Figure 8a. Time-history plots of daily sulfate and SO_2 concentrations for the 10 central monitoring stations: (a) Sulfate concentrations, (b) SO_2 concentrations. Predicted = ..., and measured = ■.

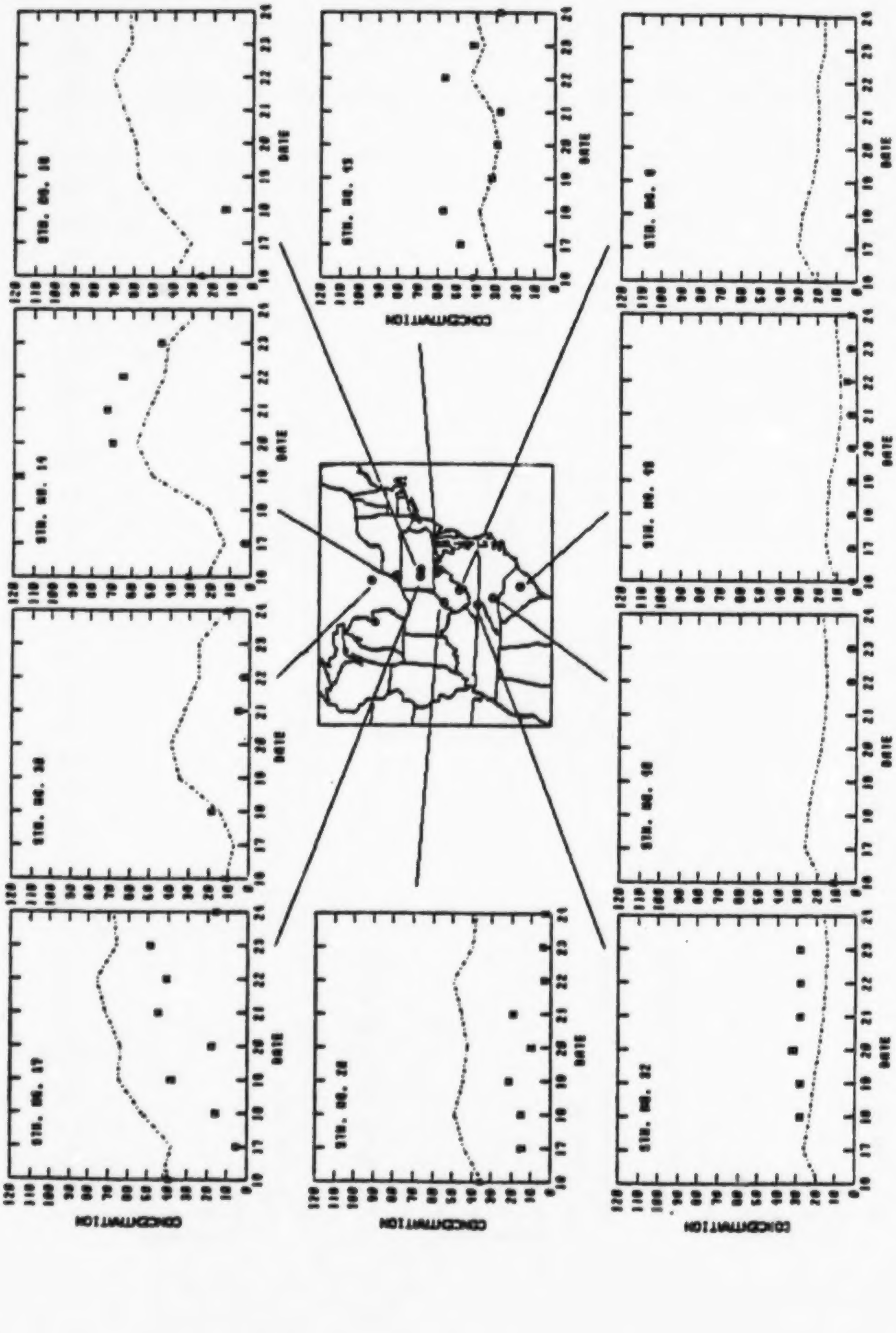


Figure 8b. (continued).

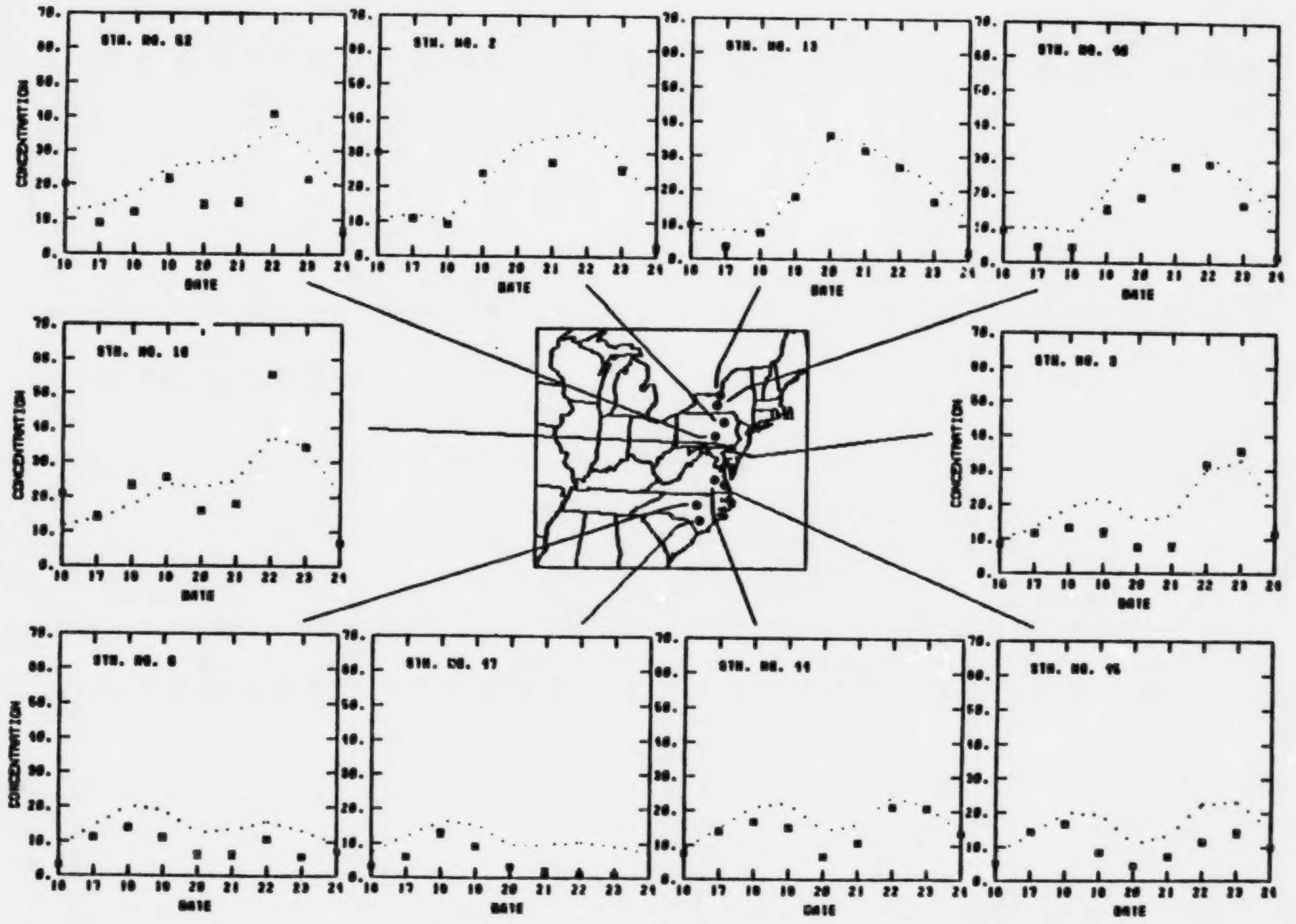


Figure 9a. Time-history plots of daily sulfate and SO_2 concentrations for the 10 easternmost monitoring stations: (a) Sulfate concentrations, (b) SO_2 concentrations. Predicted = ..., and measured = ■.

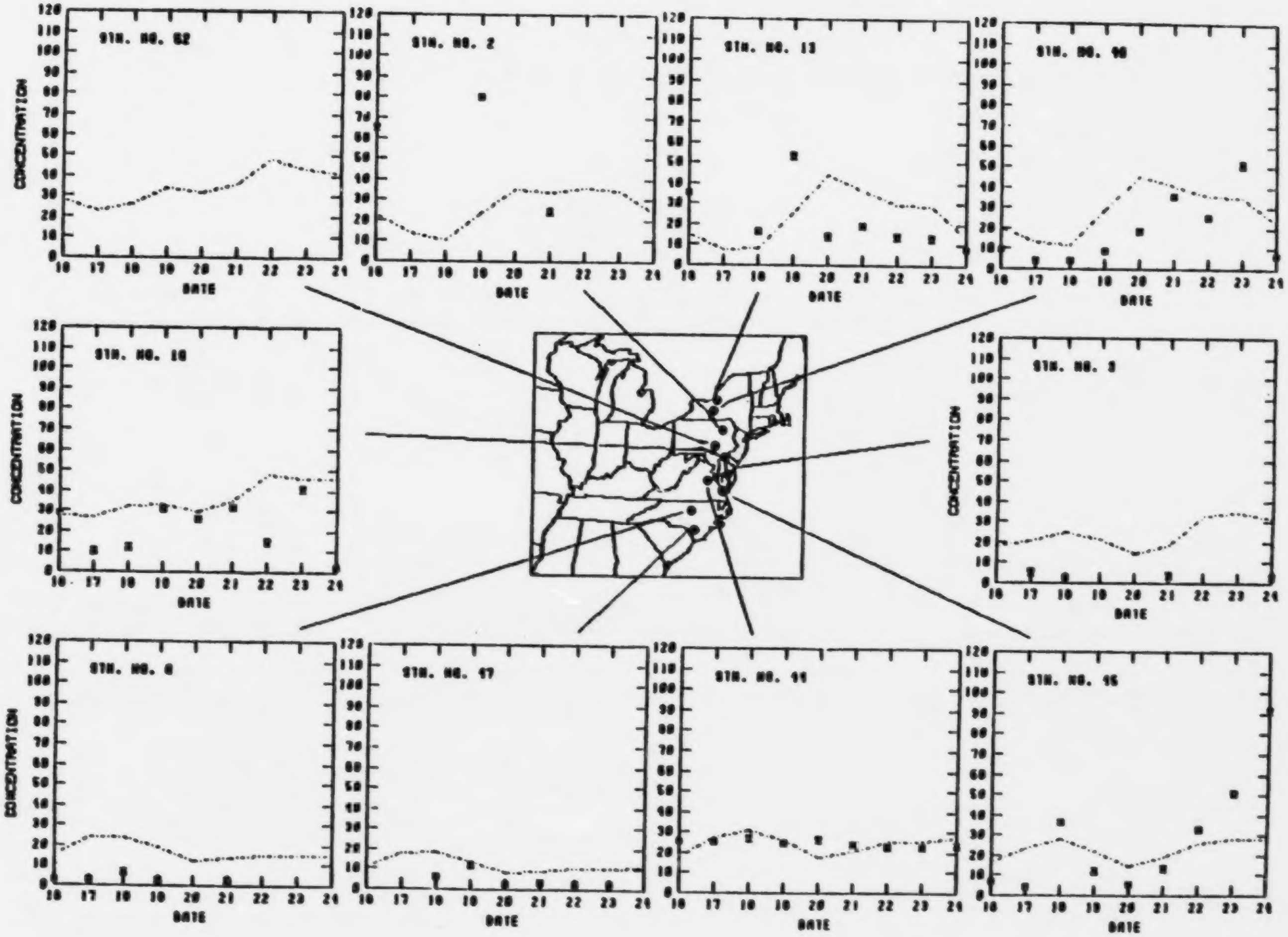


Figure 9b. (continued).

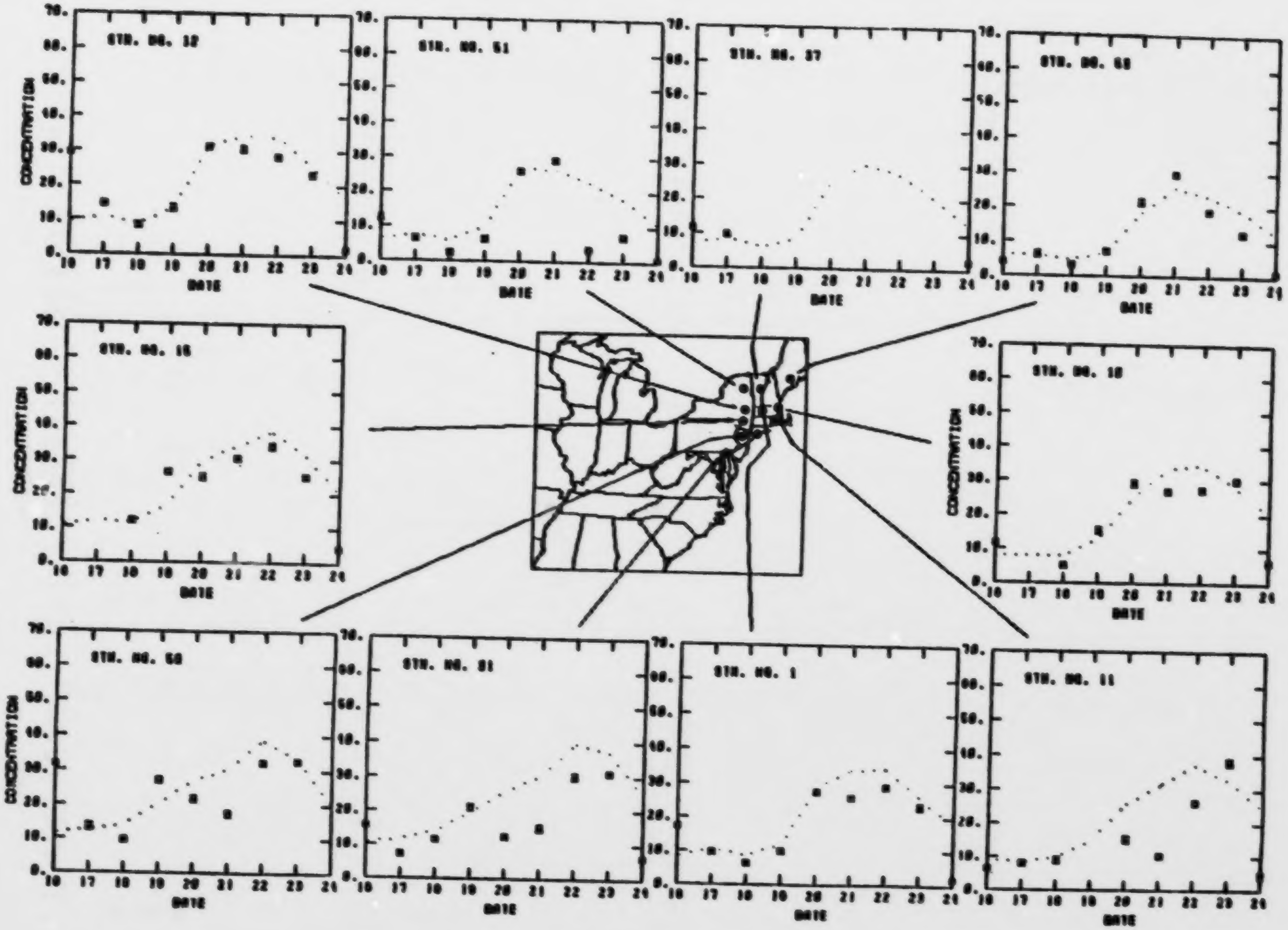


Figure 10a. Time-history plots of daily sulfate and SO_2 concentrations for the 10 easternmost monitoring stations: (a) Sulfate concentrations, (b) SO_2 concentrations. Predicted = ..., and measured = ■ .

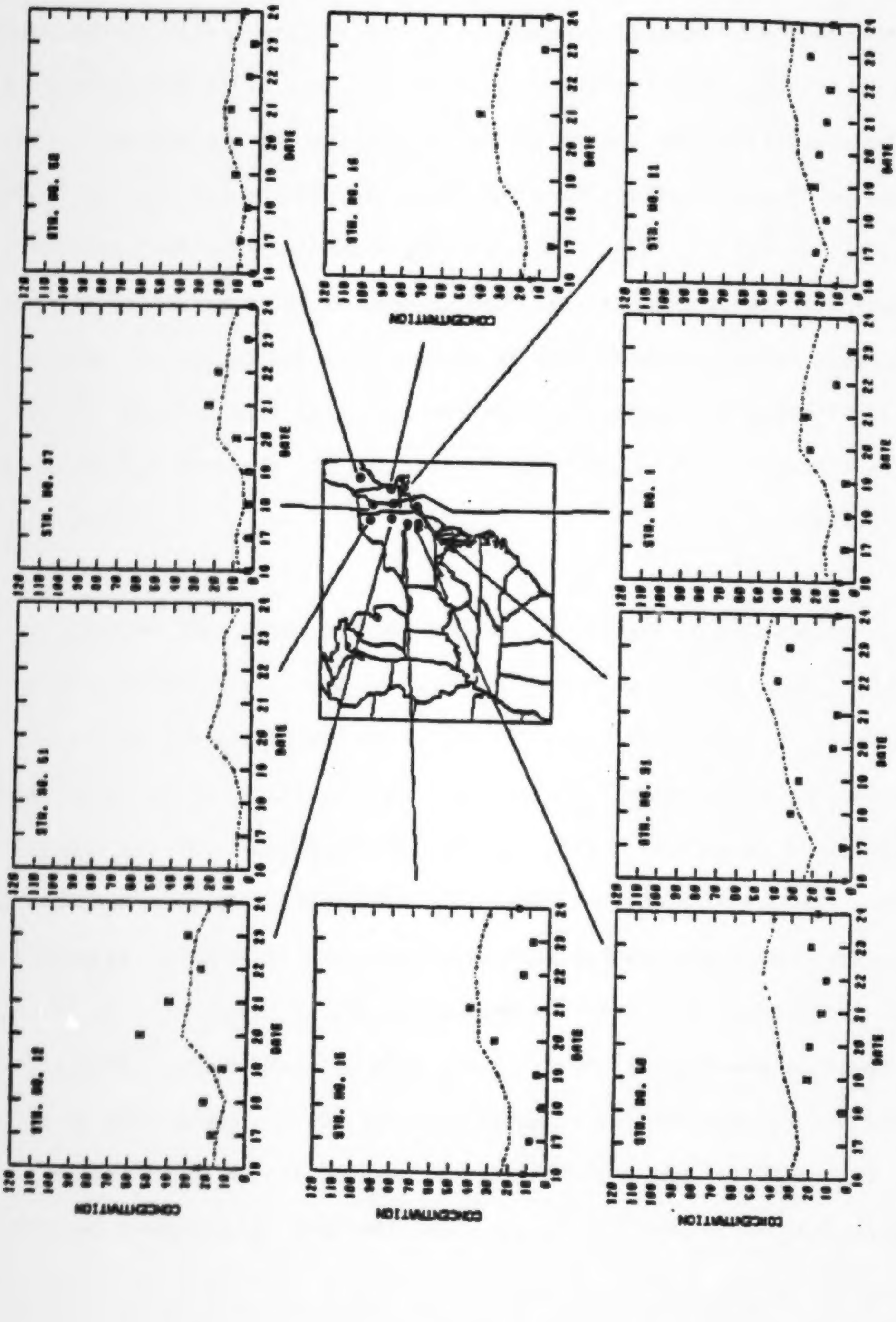


Figure 10b. (continued).

that time. Although the model does produce a slight increase in sulfate levels, the magnitude is underpredicted by more than 10 $\mu g/m^3$.

The model also appears to overpredict the sulfate minima in the northeast region on July 21, 1978 (Stations 1, 10, 11, 31, and 50 in Figure 10). A possible explanation for the overprediction of the sulfate minima at these stations may derive from the fact that they are all located near the coast. During July 20 and 21, 1978, the prevailing wind direction for air flow aloft was approximately parallel to the coast. However, an analysis of the surface weather maps of this period shows an onshore flow during daytime hours. A slight deflection of the onshore surface wind, most likely caused by the diurnal land/sea breezes, could conceivably account for the observed dip in sulfate concentrations.

An examination of Figures 6b through 10b indicates that several of the SO_2 concentration trends are well simulated by the model (e.g., Stations 12, 14, and 43), whereas in other cases, the SO_2 concentration trends are completely missed by the model. At one-fourth of the monitoring stations, there is no obvious overprediction or underprediction in SO_2 concentrations. Of the remaining stations, the majority of the predictions are higher than the observations. It is, however, not clear whether this overprediction tendency is related to the siting of the rural stations. An analysis recently carried out on hourly SO_2 concentrations observed in the St. Louis RAPS network (Hanna, 1982) suggests that the natural variability of hourly average SO_2 concentrations is greater than a factor of 2. One would expect the variability of 24-h average SO_2 concentrations to be somewhat less. Thus, the lack of agreement between the measured and predicted SO_2 concentrations cannot be considered to be solely a result of natural variability. Because SO_2 is a primary emission, its distribution should be closely associated with the distribution of major emitting sources. Thus, inadequate subgrid-scale treatment of point sources in the model could possibly lead to the discrepancies between the predicted and measured SO_2 concentrations.

Model performance statistics were computed to quantitatively assess its predictive capability. In general, the model tends to overpredict low concentrations and underpredict high concentrations. This trend is more pronounced for SO_2 than for sulfate. Nevertheless, the statistics show that sulfate predictions have an overall bias of $-2.3\ \mu g/m^3$ and a correlation coefficient of 0.80 when compared with the measurements. Similar statistics for SO_2 indicate a bias of $-8.6\ \mu g/m^3$ and a correlation coefficient of 0.42.

APPENDIX. INCORPORATION OF AN AEROSOL MODULE

General Considerations of Aerosol Dynamics and Chemistry

The environmental effects associated with aerosols at the regional scale involve the dry and wet deposition of sulfate and nitrate species, visibility impairment, and health effects. We will briefly address these three major issues before we discuss the state of the art in modeling aerosol dynamics and chemistry.

Aerosols that are formed in the atmosphere include primary aerosols that have been directly emitted into the atmosphere from natural sources (e.g., volcanoes, plants, soil, sea salt) or anthropogenic sources, and secondary aerosols that result from the gas-to-aerosol conversion of condensable chemical species that often have been formed through gas phase chemical reactions. An atmospheric aerosol population consists of three main modes that differ in concentration and relative importance according to the conditions considered: a nuclei mode (aerosols ranging from 0.001 to 0.01 μm in diameter) that corresponds to new nucleated aerosols; an accumulation mode (aerosols ranging form 0.1 to 2 μm in diameter) that corresponds to secondary aerosols formed by gas-to-aerosol conversion and growth of nuclei aerosols; and a coarse mode (aerosols larger than 2 μm in diameter) that corresponds to mechanically generated aerosols and interacts little with the other two modes. The size distribution of the aerosol population depends on the relative importance of these three modes and on the dynamic processes that affect its evolution. The chemical composition of the chemical processes that have led to secondary aerosol formation. The chemical composition is a function of the aerosol size and varies as gas-to-aerosol conversion, aerosol-phase chemical reactions, coagulation, sedimentation, deposition, emission, washout and rainout of aerosols occur. The environmental effects of aerosols depend on both aerosol size distribution and aerosol chemical composition.

Dry and wet deposition of sulfate and nitrate aerosols contribute to the acidification of soils and watersheds. A detailed treatment of these processes in a regional model may require a description of the aerosol size distribution and chemical composition. The dry deposition velocity of the aerosol will depend on its size. Wet deposition occurs when aerosols are rained out or washed out. Their initial chemical composition will determine to a certain extent the kinetics of the droplet chemistry, because many oxidation reactions are pH dependent. It is likely that rainout efficiency does not depend notably on particle size. However, washout by cloud droplets under the cloud base will be more effective for aerosols larger than 1.0 μm in diameter (removal by inertial impaction) and for aerosols smaller than 0.1 μm in diameter (removal by Brownian motion) than for aerosols in the intermediate size range of 0.1 to 1.0 μm in diameter.

The modeling of regional haze necessarily requires the modeling of the aerosol size distribution and chemical composition, because light scattering and absorption depend on these aerosol characteristics. Light scattering occurs primarily for fine aerosols in the 0.1- to 3.0-μm-diameter range. Light absorption by aerosols depends on the amount of carbonaceous aerosol present.

The health effects of aerosols are size dependent. Large aerosols will generally be deposited in the extrathoracic region, and small aerosols will be removed by Brownian motion. Aerosols in the 0.2- to 15-μm-diameter range are inhaled by humans and can be deposited in the tracheobronchial airways. Based on these considerations, the State of California Air Resources Board has issued an air quality standard for aerosols less than 10 μm in diameter, and the U.S. Environmental Protection Agency is also considering the promulgation of a standard for fine aerosols.

State of the Art in Atmospheric Aerosol Modeling

The evolution of the aerosol size distribution in the atmosphere is governed by the so-called General Dynamic Equation, which may be expressed as follows for the aerosol number distribution n (v,t):

$$\begin{aligned}\frac{\partial n}{\partial t} = &- \nabla(\underline{u}\cdot n) + \nabla K \nabla n + \frac{1}{2}\int_0^v \beta(v - \tilde{v}, \tilde{v})\, n(v - \tilde{v})\, n(\tilde{v})\, d\tilde{v} \\ &- \int_0^\infty \beta(v, \tilde{v})\, n(v)\, n(\tilde{v})\, d\tilde{v} + \frac{\partial}{\partial v}\, n\left(\frac{\partial v}{\partial t}\right) \qquad \text{(A-1)} \\ &+ S(v, t) - R(v, t).\end{aligned}$$

where v is the aerosol volume and t is the time. The term on the left-hand side represents the change in the aerosol number distribution with time. The first term on the right-hand side, where u is the wind field vector, represents advection; the second term, where K is the eddy-diffusivity tensor, represents atmospheric diffusion; the third term, where β is the coagulation of aerosols of volume $\tilde{v}$, represents production of aerosols of volume v; the fourth term represents the coagulation of aerosols of volume v; the fifth term represents the growth of aerosols by gas-to-aerosol conversion; the sixth term represents sources of aerosols by emissions or nucleation of condensable species; and the last term represents removal of aerosols by sedimentation, surface deposition, washout, and rainout.

The numerical solution of the General Dynamic Equation has been presented by Gelbard and Seinfeld (1979). It is, however, computationally expensive, and one must introduce some simplifying assumptions for atmospheric applications. Two major approaches have been considered. Eltgroth and Hobbs (1979) represented three modes of the aerosol size distribution by lognormal sulfate aerosol. The approach, introduced by Gelbard, Tambour, and Seinfield (1980), consisted of approximating the size distribution by a discrete approximation, the so-called sectional representation. This concept was extended at Systems Applications, Inc., and applied to sulfate aerosol formation (Seigneur, 1982) and nitrate aerosol formation (Seigneur, Saxena, and Hudischewskyj, 1982) in power plant plumes. Comparisons of model predictions with smog chamber and atmospheric data have been satisfactory.

These models considered the dynamics of chemically inert aerosols. The growth law of an aerosol due to chemical reaction in the aerosol phase, in addition to gas-to-aerosol conversion, has been modeled and, for example, may include a detailed treatment of the thermodynamic equilibria between the gas phase and liquid phase, diffusion-limited condensation, chemical reactions in the aerosol, and the deliquescence of ammonium nitrate or ammonium sulfates

(Saxena, Seigneur, and Peterson, 1983). This approach has been applied to the chemistry of cloud or fog droplets (e.g., Seigneur, Saxena, and Roth, 1983; Jacob and Hoffmann, 1983). The absorption of gases by falling raindrops below the cloud base has also been considered in theoretical studies (Adewuji and Carmichael, 1982; Reda and Carmichael, 1982; Durham, Overton, and Aneja, 1981).

The coupling of aerosol dynamics and chemistry can be achieved by solving Equation (15) when the growth law term includes the growth due to chemical reaction in the aerosol phase. The formulation of such a model has been presented by Bassett, Gelbard, and Seinfeld (1981).

Regional Modeling of Aerosols

Until now, modeling of sulfate and nitrate formation in regional models has been limited to parameterized representations (e.g., pseudo-first-order oxidation of SO_2 to sulfate). However, the use of modeling to evaluate emission control strategies will require a more accurate description of the nonlinear chemical and physical processes involved.

The incorporation of aerosol dynamics and chemistry into a regional model appears, therefore, necessary. This model development effort must take into account many interacting modules. First, it is necessary to couple the gas-phase and liquid-phase chemistry. In present models, liquid-phase chemistry is assumed not to affect gas-phase chemistry. Then, it is necessary to introduce the effect of aerosol chemistry in the General Dynamic Equation according to the formulation advanced by Gelbard and Seinfeld (1980). In parallel to the model development effort, model evaluation studies must also be continuously conducted to assure that the modeling approach is correct. These efforts are presently underway at Systems Applications, Inc.

The computational requirment of a regional-scale model will necessitate the simplification of the aerosol dynamics and chemistry module. This must be done carefully in order to maintain an adequate balance between reasonable computational costs and model prediction accuracy. The simplifying assumptions that can be made will allow to reduce the complexity of the General Dynamic Equation. For instance, the number of aerosol size sections that constitute the size distribution can be chosen to be minimal while still preserving the calculational accuracy. Coagulation may generally be neglected for regional-scale aerosol concentrations, since other processes such as gas-to-aerosol conversion are likely to prevail. The chemical kinetic mechanism for the aerosol phase may be limited to only the most important chemical reactions.

The result of the incorporation of aerosol dynamics and chemistry into a regional model will be a model that can provide quantitative information on sulfate, nitrate, and cloud pH, while taking into account the major chemical and physical processes that govern the formation of acidic species in the atmosphere.

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STEM MODEL FOR THE REGIONAL TRANSPORT OF PHOTOCHEMICAL OXIDANTS AND THEIR PRECURSORS*

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INTRODUCTION

The relationships between emissions and the distribution of photochemical oxidants and their precursors are complex and not fully understood. Many trace species may be transported long distances (tens to thousands of kilometers) from source areas, resulting in high ambient levels over broad regions. In addition, many of these compounds have been related to a number of environmental problems, including acid rain, tropospheric haze, and reduced visibility, and they have been correlated with a variety of adverse health indicators.

The distribution of these species in the atmosphere is the result of complex interactions among emissions (anthropogenic and natural), prevailing meteorology, chemical transformation, and removal mechanisms. Regional-scale transport/chemistry models that describe the circulation of photochemical oxidants and their precursors in the troposphere can be extremely beneficial in understanding the physical and chemical processes occurring between the sources and sinks of these pollutants.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

In this paper, a combined transport/chemistry model for the regional-scale transport of photochemical oxidants and their precursors is described. Called STEM (sulfate transport emissions model), the model is Eulerian and three-dimensional, and is an extension of an operational SO_x transport model developed by the authors (Carmichael and Peters, 1981). This second-generation model handles 40 species, 19 of which are advected. The advected species are NO, NO_2, SO_2, SO_4^{2-}, O_3, HNO_3, NH_3, PAN, H_2O_2, HCHO, HC_1 (alkanes), C_2H_4, HC_2 (alkenes), HC_3 (aromatics), RCHO, ROOH, HNO_2, $RONO_2$, and RO_2NO_2. Both heterogeneous and homogeneous chemical reactions and wet and dry removal processes are modeled. A finite-element numerical method is used in the model. Model formulation and initial test results are presented in the following section.

MODEL DESCRIPTION

The regional transport of photochemical oxidants and their precursors is modeled within an Eulerian framework. A block description of the model is presented in Figure 1. Forty chemical species are included in the analysis. Nineteen of them are sufficiently long lived under certain circumstances that they must be treated as advected species. The remaining species are short lived and are modeled by steady-state methods. The mathematical model contains no regional or area-specific information, and the

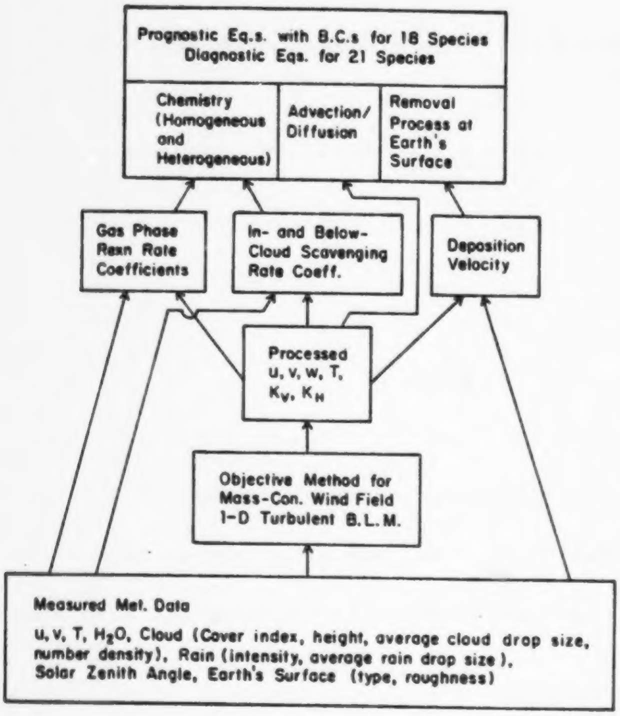

Prognostic Eq.s. with B.C.s for 18 Species
Diagnostic Eqs. for 21 Species
Chemistry (Homogeneous and Heterogeneous)
Advection/ Diffusion
Removal Process at Earth's Surface
Gas Phase Rexn Rate Coefficients
In- and Below-Cloud Scavenging Rate Coeff.
Deposition Velocity
Processed u, v, w, T, K_V, K_H
Objective Method for Mass-Con. Wind Field 1-D Turbulent B.L.M.
Measured Met. Data
u, v, T, H₂O, Cloud (Cover index, height, average cloud drop size, number density), Rain (intensity, average rain drop size), Solar Zenith Angle, Earth's Surface (type, roughness)

Figure 1. Schematic of model construction.

number of grids and the grid spacing can be chosen for a particular application. The mathematical analysis is based on the coupled, three-dimensional advection-diffusion equation:

$$\frac{\delta C_\ell}{\delta t} + \frac{\delta(U_i C_\ell)}{\delta x_i} = \frac{\delta}{\delta x_j}\left[K_{jj}\,\frac{\delta C_\ell}{\delta x_j}\right] + R_\ell + S_\ell , \qquad (1)$$

where C_ℓ is the concentration of species ℓ, U_i is the velocity vector, K_{jj} is the eddy diffusivity tensor ($K_{ij}=0$ for $i \neq j$ has been assumed), R_ℓ is the rate of formation or loss by chemical reaction, and S_ℓ is the emission rate.

The model actually utilizes the surface-following coordinate system shown in Figure 2. The vertical region, including topographical features, is mapped into a dimensionless rectangular region according to:

$$\rho_k = \frac{Zk - h(x,y)}{H_2\ (x,y,t)}\ h(y) \leq Z_k \leq h(x,y) + H_2(x,y,t), \qquad (2)$$

and

$$\rho_k = \left(\frac{k - 1}{\mathrm{KGRID-1}}\right)^{\alpha}\ 0 \leq \rho_k \leq 1,\ k = 1,\ \mathrm{KGRID} \qquad (3)$$

where k (subscript) = the vertical grid number,
KGRID = the total number of grids, and
α = a parameter controlling the grid spacing.

When $\alpha = 1$, the grid spacing in the dimensionless coordinate is uniform; when $\alpha > 1$, there is higher resolution near the surface. Our current application uses $\alpha = 2$ and $H_2(x,y,t) = 8$ km. The height of the region is chosen so that boundary-layer-free troposphere processes and interactions can be modeled.

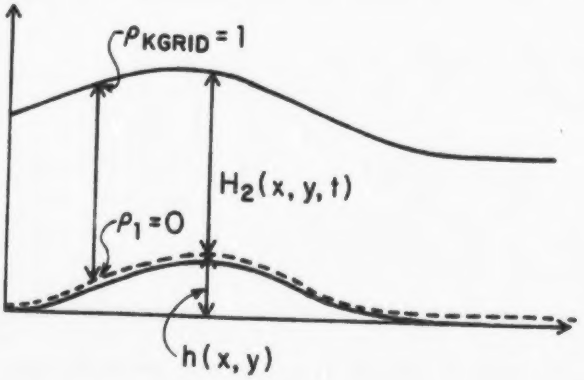


Figure 2. Suface-following coordinate system used in the model.

Chemistry

Both gas-phase and liquid-phase chemical reactions are treated in the model. The homogeneous gas-phase mechanism used is summarized in Table 1. The mechanism involves 84 reactions and 40 species. NO, NO_2, HNO_3, NH_3, SO_2, SO_4^{2-}, HC_1 (alkanes), C_2H_4, HC_2 (alkenes), HC_3 (aromatics), O_3, PAN, HCHO, RCHO, H_2O_2, $RONO_2$, and RO_2NO_2 are treated as transported species, and the remainder are treated as pseudo-steady-state species. The extensiveness of the chemical mechanism enables the modeling of urban chemistry as well as nonurban tropospheric chemistry.

The model's treatment of the chemistry also includes the interactions between the gas-phase chemistry and the heterogeneous removal processes. Thus, additional reactions of the form noted below are added to the gas-phase chemical mechanism. Two types of heterogeneous removal processes are treated in the model: wet removal (both in-cloud and below-cloud) and deposition on aerosol surfaces, i.e.,

$$C_{i_{(g)}} \xrightarrow{k_{iwet}} C_{i_{(\ell)}} + \text{Products}, \qquad (4a)$$

and

$$C_{i_{(g)}} \xrightarrow{k_{ihet}} C_{i_{(a)}} + \text{Products}, \qquad (4b)$$

TABLE 1. HOMOGENEOUS GAS-PHASE KINETICS MECHANISM USED IN THE STEM MODEL

---

1. $NO_2 + hv - NO + O\ (^3P)$
2. $O\ (^3P) +)_2 + M - O_3 + M$
3. $O_3 + NO - NO_2 + O_2$
4. $NO_2 + O\ (^3P) - NO + O_2$
5. $NO_2 + O\ (^3P) - NO_3$
6. $NO + O\ (^3P) - NO_2$
7. $NO_2 + O_3 - NO_3 + O_2$
8. $NO_3 + NO - 2\ NO_2$
9. $NO_3 + NO_2 - N_2\ O_5$
10. $N_2O_5 - NO_2 + NO_3$
11. $N_2O_5 + H_2O - 2HONO_2$
12. $NO + NO_2 + H_2O - 2HONO$
13. $HONO + HONO - NO + NO_2 + H_2O$
14. $O_3 + h\nu - O_2 + O\ (^1D)$
15. $O_3 + h\nu - O_2 + O\ (^3P)$
16. $O(^1D) + M - O\ (^3P) + M$
17. $O\ (^1D) + H_2O - 2OH$
18. $HO_2 + NO_2 - HONO + O_2$
19. $HO_2 + NO_2 - HO_2NO_2$
20. $HO_2NO_2 - HO_2 + NO_2$
21. $HO_2 + NO - NO_2 + OH$
22. $OH + NO - HONO$
23. $OH + NO_2 - HONO_2$
24. $HONO + h\nu - OH + NO$
25. $CO + OH - CO_2 + HO_2$
26. $OH + HONO - H_2O + NO_2$
27. $HO_2 + HO_2 - H_2O_2 + O_2$
28. $H_2O_2 + h\nu - 2OH$
29. $OH + HO_2 - H_2O + O_2$
30. $OH + O_3 - HO_2 + O_2$
31. $HO_2 + O_3 - OH + 2O_2$
32. $HCO + h\nu - HO_2 + HCO$
33. $HCO + h\nu - H_2 + CO$
34. $HCHO + OH - HCO\ (+H_2O)$
35. $HCO + O_2 - HO_2\ (+CO)$
36. $RCHO + h\nu - RO_2 + HCO$
37. $RCHO + OH - RCO_3$
38. Ol (olefin) $+ OH - RO_2$
39. $Ol + O - RO_2 + RCO_3$
40. $Ol + O_3 - 0.5\ \zeta\ HCHO + (1 - 0.5\ \zeta)\ RCHO$
    $+0.5\ (\epsilon)\ (\xi + 0.5\ \eta)\ RO_2$
    $+0.25\ (\epsilon)(\xi + \eta)\ HO_2 + 0.25\ (\epsilon\xi)OH$
    $+0.5\ (1 - \epsilon)\ RCHOO$
    $+0.25\ (\epsilon\eta)\ RO + 0.25\ (\epsilon\eta)\ HCO$
41. $Alk + OH - RO_2$
42. $Alk + O - RO_2 + OH$
43. $C_2H_4 + OH - RO_2$
44. $C_2H_4 + O - RO_2 + HCO$
45. $RO - \alpha\ HO_2 + (1 - \alpha)\ RO_2 + \beta\ HCHO + \gamma\ RCHO$
46. $NO + RO - RONO$
47. $RONO + h\nu - RO + NO$
48. $NO_2 + RO - RONO_2$
49. $NO_2 + RO - RCHO + HONO$
50. $NO_2 + RO_2 - RO_2NO_2$
51. $NO_2 + RO_2 - RCHO + HONO_2$
52. $RO_2\ NO_2 - NO_2 + RO_2$
53. $NO + RO_2 - NO_2 + RO$
54. $NO + RCO_3 - NO_2 + RO_2$
55. $NO_2 + RCO_3 - PAN$
56. $PAN - NO_2 + RCO_3$
57. $Aro + OH - RO_2 + RCHO$
58. $SO_2 + HO_2 - HO + SO_3$
59. $SO_2 + HO - HSO_3$
60. $SO_2 + O\ (^3P) - SO_3$
61. $SO_2 + RO_2 - RO + SO_3$
62. $SO_2 + RO - R\ SO_3$
63. $SO_2 + RCHOO - RCHO + SO_3$
64. $SO_2 - Part$
65. $SO_3 + H_2O - H_2\ SO_4$ (Part)
66. $HSO_3 + O_2 - HSO_5$
67. $HSO_5 + H_2O - HSO_5\ (H_2O)$ (Part)
68. $RSO_3 + O_2 - RSO_5$
69. $RSO_5 + H_2O - RSO_5\ (H_2O)$
70. $RCHOO + H_2O - RCOOH + H_2O$
71. $NH_3 + OH - NH_2 + H_2O$
72. $NH_2 + O_2\ {}^{M}\ NH_2O_2$
73. $NH_2\ O_2 + NO - NH_2O + NO_2$
74. $NH_2\ O_2 + OH - NH_2\ OH + O_2$
75. $NH_2O + O_2 - HNO + HO_2$
76. $HNO + OH - NO + H_2O$
77. $HNO + O_2 - NO + HO_2$
78. $NH_3 + HNO_3\ {}^{M}\ NH_4\ NO_3$
79. $HNO_3 + OH - NO_3 + H_2O$
80. $HNO_3 + h\nu - NO_2 + OH$
81. $NO_3 + h\nu - NO_2 + O$
82. $RO_2 + HO_2 - ROOH + O_2$
83. $ROOH + n - RO + HO$
84. $ROOH + OH - RO_2 + H_2O$

where k_{iwet} = the wet removal coefficient, and

k_{ihet} = the removal coefficient for deposition on aerosol surfaces.

The wet removal coefficient, k_{iwet}, is calculated in the model from a parameterization based on the liquid water content of the cloud, cloud temperature, characteristic droplet size, number density of cloud droplets, cloud pH and rainfall intensity, and the chemical and physical properties of the absorbed species (i.e., Henry's Law constant, gas-phase diffusivity, and dissociation and redox reactions in solution). Also included is the liquid-phase generation of species like sulfate. The sulfate production rate in clouds, which is calculated by using the above parameters, is based on the reactions S(IV) + O_3 and S(IV) + H_2O_2. (See Hong and Carmichael, 1982, for a complete description of the calculation of k_{iwet}). The wet removal rates for some soluble species, calculated by the above procedure for in-cloud conditions with a cloud temperature of 0°C, a droplet size of 30 μm, a pH of 4.7, and a liquid water control of 1.5 g/m^3, are summarized in Table 2. The rates reflect the efficiency with which cloud droplets remove these highly soluble species.

The removal coefficient for deposition on aerosol surfaces is calculated by using classical mass transfer theory and gas kinetic theory. It is described in detail by Luther and Peters (1982). In this treatment, the aerosol particles are assumed to be spherical and are described by the physical and chemical properties of water; desorption phenomena are not included. Values of k_{ihet} for selected compounds, calculated by using the aerosol residence time distribution of Jaenicke (1978) and the aerosol size distribution of Junge (1963) with a

TABLE 2. IN-CLOUD REMOVAL RATES FOR SOLUBLE SPECIES CALCULATED BY THE PARAMETERIZATION PROCEDURE DISCUSSED IN THE TEXT

Species	k_{wet} (s^{-1})
HNO_3	0.06
NH_3	0.102
SO_2	0.28×10^{-4}
SO_4^{2-}	-0.28×10^{-4} (production rates)
PAN	0.018
HCHO	0.086
RCHO	0.071
H_2O_2	0.088
HNO_2	0.07

surface area of 520 $\mu m^2/m^3$, and an accommodation coefficient of 1, are presented in Table 3. These values represent upper limits.

The chemical reaction rate constants are calculated in the model and vary with temperature and photon flux. The effects of clouds on the photon flux are included in the calculation by using an empirical correlation relating photon flux to cloud cover (Kaiser and Hill, 1976); i.e.,

$$G = G_c \, (1 - A \, C^{1.75}), \qquad (5)$$

TABLE 3. CALCULATED HETEROGENEOUS LOSS CONSTANTS FOR AEROSOL/GAS INTERACTIONS

Species	k_{het} (s^{-1})
NO	1.4×10^{-6}
O_3	6.7×10^{-6}
OH	1.1×10^{-1}
H_2O_2	6.2×10^{-2}
HNO_3	6.5×10^{-2}
NO_3	6.7×10^{-2}
O_2	7.0×10^{-7}
NO_2	7.6×10^{-2}

where G_c = the photon flux for clear sky,

C = the cloud cover fraction, and

A = a constant dependent on cloud type and has a value of 0.55 for fair-weather cumulus clouds or patches of cirrus or cumulus clouds.

Furthermore, the enhancement of photon flux due to cloud albedo is included in the analysis by multiplying the clear-sky flux at grid points directly above the cloud level by a factor of 1.3.

Numerics

The simulation of regional transport/chemistry described by Equation (1) requires numerical integration. The method used in the model is a combination of the concept of fractional time steps and that of one-dimensional finite elements. This is referred to as the Locally-One-Dimensional, Finite-Element Method (LOD-FEM). The LOD procedures (Mitchell, 1969; Yanenko, 1971) split the multidimensional partial differential equation into time-dependent, one-dimensional problems, which are solved sequentially.

The time-split equations are of the form:

$$\frac{\delta C_\ell}{\delta t} + L_x C_\ell = 0 \qquad (6)$$

$$\frac{\delta C_\ell}{\delta t} + L_y C_\ell = 0, \qquad (7)$$

$$\frac{\delta C_\ell}{\delta t} + L_z C_\ell = S_\ell . \qquad (8)$$

and

$$\frac{\delta C_\ell}{\delta t} = R_\ell , \tag{9}$$

where the L's represent the one-dimensional operators (e.g., $L_x C_\ell = \delta(U\ C_\ell)/\delta X - \delta/\delta x\left(K_{xx}\ \delta C_\ell/\delta x\right))$.

The chemical reaction term is treated separately, because many of the reactions have time scales much smaller than that for the transport. Splitting out the reaction term allows different time steps to be used for the transport and the chemistry processes. The time steps used in current applications are 0.1 min for the chemistry equations and 15 min for the transport.

The transport equations are solved by a modified Galerkin finite element method (FEM) using asymmetric weighting functions and the Crank-Nicholson approximation for the time derivative. The chemistry equations are solved by a pseudo- linearization procedure, which gives analytical approximations to the equations. The solution procedures for the transport and chemistry are discussed in more detail below.

Solution of the Chemistry Equations--

The technique used to integrate the chemistry of the transported species is an adaption of the semi-implicit Euler method proposed by Preussner and Brand (1981). With this technique, Equation (9) can be written in the following form:

$$\frac{dC_\ell}{dt} = -C_\ell \sum_j d_\ell{}^j \prod_{k \neq \ell} C_k + \sum_i p_\ell{}^i \prod_m C_m, \tag{10}$$

where $d_\ell{}^j$ = the rate constant of the destruction of species ℓ by reaction j,

$p_\ell{}^i$ = the rate constant of the production of species ℓ by reaction i,

k and m = the reactant species involved in the destruction and production reactions.

Under the assumption that all species concentrations except ℓ are known (either at the previous time) or have just been calculated, Equation (10) is of form $dC_\ell/dt + DC_\ell = P$, which has the analytic solution:

$$C_\ell \Big|_{t_i = \Delta t_i} = \frac{P}{D}\Big|_{t_i} + \left(C_\ell - \frac{P}{D}\right)\Big|_{t_i} e^{-D\Delta t_i}, \tag{11}$$

where $C_\ell |_{t_i}$ at t = 0 is the initial concentration of species ℓ. This procedure has the important physical properties that C_ℓ can never become negative provided that the reaction rates are positive and the initial values are nonnegative, and that in the limit, as $t \to \infty$, the proper equilibrium concentrations are obtained, i.e.,

$$C_\ell^{eq} = \frac{P}{D} = \frac{\sum\limits_i P_\ell^{i} \prod\limits_m C_m}{\sum\limits_j d_\ell^{j} \prod\limits_{k \neq \ell} C_k} . \qquad (12)$$

The above equation is used in the model to calculate only the advected species. However, terms P and D also contain the short-lived species (e.g., OH, HO_2, etc.). These species are calculated by using the pseudo-steady-state approximations. The use of these equations results in a set of algebraic equations that depend on the advected species concentrations. The calculation procedure to advance from t_i to $t_i + \Delta t_i$ uses $C_\ell \mid t_i$ to calculate the short-lived species concentrations and then uses these concentrations to advance $C_\ell \mid t_i$ to $C_\ell \mid t_i + \Delta t_i$ using Equation (11).

The accuracy of this technique depends on the species of interest and Δt_i. For long-lived species (e.g., SO_2, CO, and CH_4), relatively large Δt_i can be used, but for relatively reactive species, much smaller time steps are required. Figure 3 displays afternoon NO_2 concentrations, calculated by using the kinetic mechanism summarized in Table 1. The results obtained by using CSMP, the semi-implicit Eulerian method (Preussner and Brand, 1981), and the "analytic" method just described are compared. After a 5-h simulation time, the (the time step used to solve the transport equations), the results differed by less than 1%. The analytic method with the free radical solver can execute faster than CSMP by a factor of 2.

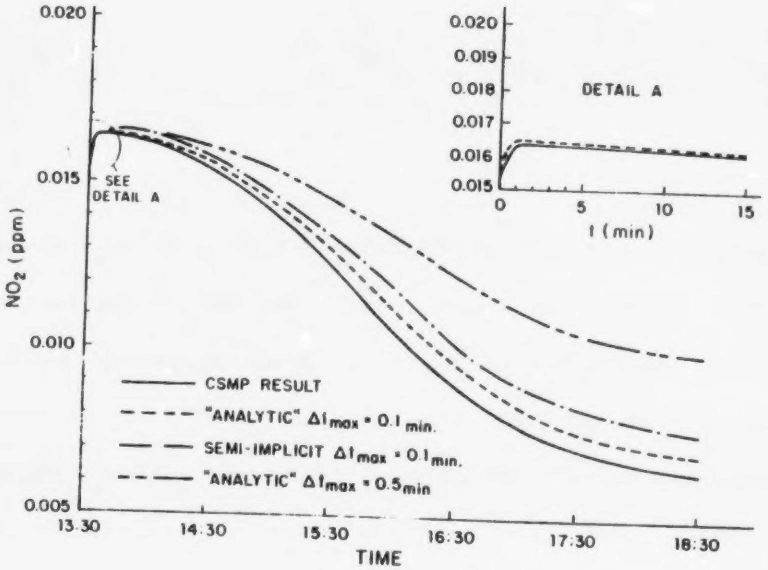


Figure 3. Afternoon NO_2 concentrations calculated by using the kinetic mechanism shown in Table 1.

Solution of the Transport Equations--

The transport equations with the boundary conditions described below are solved by using a Crank-Nicolson/Galerkin finite element method with piecewise linear trial functions, asymmetric weighting functions, and a filter to eliminate high-frequency numerical noise. Selection of the finite element method was based on the results of one- and two-dimensional numerical experiments comparing available numerical methods (Carmichael et al., 1980; Chock and Dunker, 1982; Pepper et al., 1980). The filter used is described by McRae et al. (1982).

The boundary conditions used in the model at the earth's surface and at the upper boundary are, respectively:

$$-\underline{\underline{K}}\ \nabla C_\ell) \cdot n_h = Q_\ell - V_{d,\ell} C_\ell \qquad (13)$$

and

$$(VC_\ell - \underline{\underline{K}}\ \nabla C\ ) \cdot n_H = F_\ell\ , \qquad (14)$$

where $\underline{\underline{K}}$ = a diagonal matrix of eddy diffusivities;

Q_ℓ = the surface flux of species ℓ ;

$V_{d,\ell}$ = the dry deposition velocity;

V = the wind velocity;

n_h and n_H = upward unit vectors normal to the earth's surface and the upper boundary, respectively; and

F_ℓ = the prescribed flux.

Stratospheric inputs of NO_x, HNO_3, O_3, etc., are handled in the model by Equation (14). The prescribed fluxes can vary both spatially and temporally.

The horizontal boundary conditions are:

$$(\text{if } n_b \cdot V \geq 0)\ (VC_\ell - \underline{\underline{K}}\ \nabla C\ ) \cdot n_b \Big|_{n+1} = (VC_\ell - \underline{K}\ \nabla C\ )\ n_b \Big|_{n} \qquad (15)$$

and

$$(\text{if } n_b \cdot V < 0)\ (VC_\ell - \underline{\underline{K}}\ \nabla C_\ell) \cdot n_b \Big|_{n+1} = (VC_{o,\ell} - \underline{\underline{K}}\ \nabla C_\ell) \cdot n_b \Big|_{n} \qquad (16)$$

where n_b = the outward unit vector normal to horizontal boundary,

subscript n = time t_n or the numerical calculation, and

$C_{o,\ell}$ = the concentration of species ℓ at the outside of the calculation region.

Test calculations of the transport equations are described in detail elsewhere (Kitada et al., 1983; Carmichael et al., 1982). These tests demonstrate that this method is efficient and stable for large wind velocities and local peclet numbers.

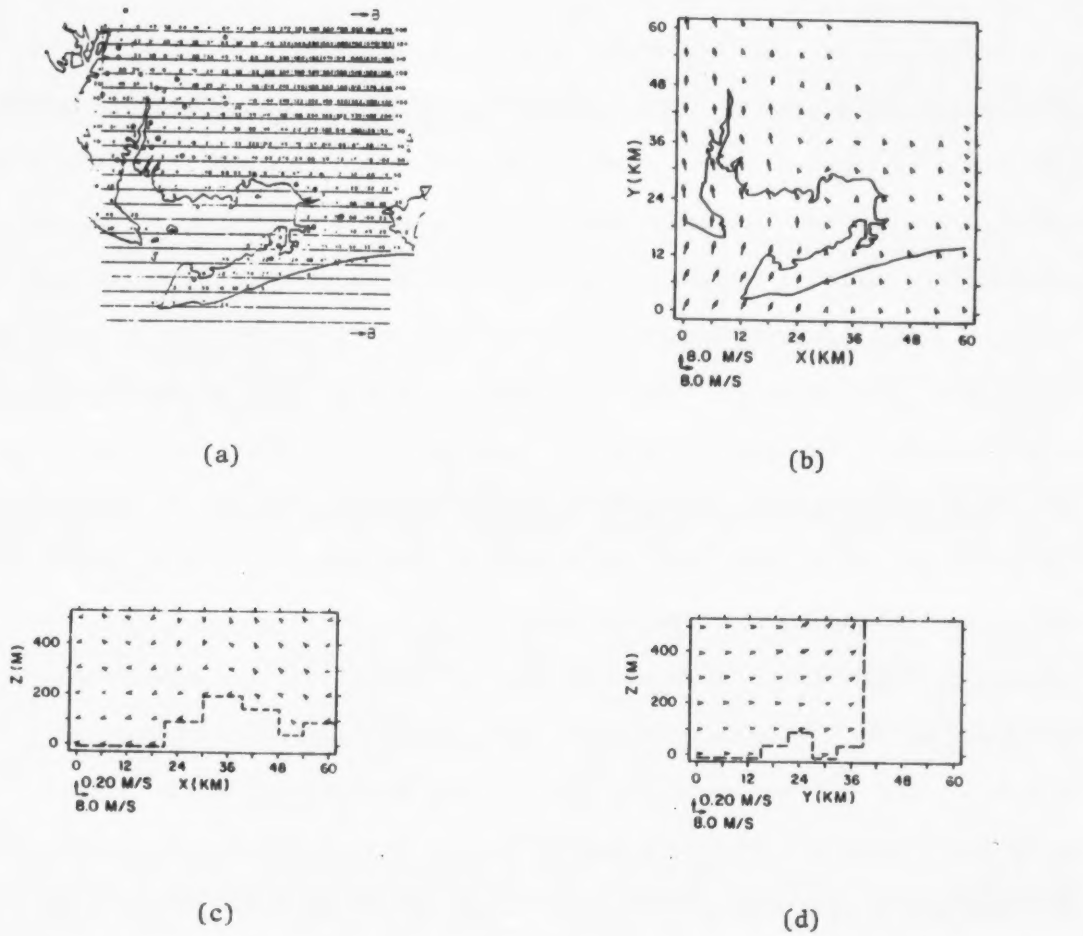
Model Inputs

The model requires the following inputs: source distributions, initial conditions, and meteorological data. The required meteorological data are summarized in Figure 1. Generally, upper air meteorological data are available at 12-h intervals. However, the boundary-layer dynamics, especially over land, have substantial temporal variation and a 24-h periodicity. Therefore, the upper air data has to be analyzed to provide the necessary temporal and spatial structure to the input data. This is very important in calculating vertical winds and eddy diffusivity values.

Objective Analysis of Wind Field--

Typically, only the horizontal wind fields are measured, and these measurements do not satisfy the continuity equation. A non-mass-conservative wind field introduces an artificial pseudo-first-order loss or generation term into the species mass transport equation (Kitada et al., 1982). In this model, an objective analysis procedure based on variational calculus is being used to obtain a three-dimensional mass-consistent wind field (Sasaki, 1970; Peters et al., 1979). Kitada et al. (1983) has applied this analysis to the Mikawa Bay area in Japan. The area, shown in Figure 4 is 60 km x 60 km. In this particular application, a vertical region of 500 m was chosen. As shown, the northwestern part of the region is blocked by mountains that extend beyond the 500-m vertical region. For the numerical calculations, the region was divided into 20 x 20 horizontal grids and 10 vertical grids. Surface wind data were available at the 27 observation locations, shown by the solid circles in Figure 4. The initial horizontal surface wind was interpolated by using a weighting factor of $1/r^2$ with a maximum radius of influence equal to twice the average separation distance between the observations points. The initial upper winds were obtained by extrapolating the surface data with a power law profile.

The derived winds at 1500 h local time on July 4, 1973, are shown in Figure 4, as well as the horizontal wind field at 200 m above sea level and the vertical wind fields along the A-A and B-B cross sections. These results demonstrate that this analysis can reproduce reasonable three-dimensional flow fields with substantial topographic features from limited horizontal input data. This analysis is currently being used to generate regional-scale flow field.



(a) (b)

(c) (d)

Figure 4. Three-dimensional wind fields for Mikawa Bav, Japan, based on the objective analysis procedure. (a) 20 x 20 horizontal grids, 27 observation points. (b) horizontal wind field at 200 m above sea level. (c) vertical windfield along A-A cross section, (d) vertical wind field along B-B cross section.

Dry Deposition Velocities--

Dry deposition velocities are calculated from estimates of SO_2 aerodynamics and surface resistances that are based on surface wind speed, surface roughness, surface evaporation rate, and stability (Carmichael and Peters, in press). The calculated values vary both temporally and spatially. The averaged calculated value for the Eastern United States for the period July 4 to July 10, 1974 (average includes day and night values over 864 grid points) is $V_dSO_2 = 0.44$ cm/s (at 20 to 30 m). Deposition velocity values for the other trace gases are obtained by scaling the SO_2 deposition velocity by a factor determined by measured deposition velocities, estimated aerodynamic resistance, and Henry's Law constants. Table 4 presents calculated values for selected species under conditions when the dry deposition velocity of SO_2 is 0.5 cm/s.

Mixing Layer Analysis--

The distribution of trace gases in the atmosphere is greatly influenced by the dynamic behavior of the boundary layer. In this model, the one-dimensional boundary-layer model developed by Yamada and Mellor (1975) is used to describe the diurnal boundary layer (specifically used to calculate K_v profiles). Examples of vertical eddy diffusivity profiles generated by this analysis are shown in Figure 5. This model can include large-scale meteorological features

TABLE 4. CALCULATED DRY DEPOSITION VELOCITIES OF SELECTED SPECIES FOR CONDITIONS WHERE V_dSO_2 = 0.5 cm/s

Species	Dry Deposition Velocity (cm/s)	Species	Dry Deposition Velocity (cm/s)
NO	0.01	HC_3 (aromatics)	0.01
NO_2	0.01	O_3	0.01
HNO_3	1.0	PAN	0.80
NH_3	0.67	HCHO	0.52
SO_4^{2-}	0.2	H_2O_2	1.0

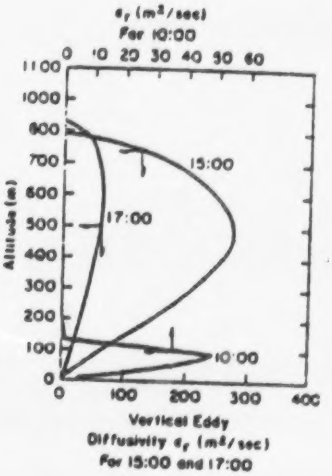


Figure 5. Example of vertical eddy diffusivity profiles from the one-dimensional turbulent boundary-layer model of Yamada and Mellor (1975).

(i.e., mesoscale or synoptic-scale) through a geostrophic wind term. The one-dimensional calculation is performed at each grid point, but it is still considerably less expensive than a three-dimensional boundary-layer model.

RESULTS AND DISCUSSION

The STEM model for the regional transport of photochemicals is currently undergoing extensive testing. A one-dimensional (Z,t) version that includes all processes except horizontal transport is being used to access the effects and importance of the various tropospheric processes (i.e., heterogeneous removal, homogeneous chemistry, stratospheric input, etc.) on the distribution of trace gases in the troposphere.

Table 5 shows predicted O_3 and OH values at selected times and elevations for the following cases:

- Without heterogeneous removal, with stratospheric sources, surface sources excluding HC sources and no clouds;
- With heterogeneous removal, with stratospheric sources, surface sources excluding HC sources and no clouds;
- With heterogeneous removal, with stratospheric sources, surface sources including HC sources and no clouds; and
- With heterogeneous removal, with stratospheric sources, surface sources including HC sources and clouds.

As shown, the O_3 and OH concentrations decrease when heterogeneous removal is included in the model (see runs 1 and 2). Without heterogeneous removal, there is a higher concentration of the soluble species (e.g., H_2O_2, HNO_3, HCHO,

TABLE 5. CALCULATED O_3 AND OH CONCENTRATIONS FOR VARIOUS CONDITIONS USING A ONE-DIMENSIONAL VERSION OF STEM

Conditions	Time (h)	Case[a]			
		1	2	3	4
O_3 at surface (ppb)	0700	17	17	22	16
	1200	58	23	63	42
	1800	176	13	75	43
	2400	~0	-	~0	~0
O_3 at 1.25 km (ppb)	0700	31	31	31	31
	1200	65	31	49	41
	1800	205	26	86	24
OH at surface (molecules/cm^3)	0700	5.4(3)	3.2(3)	1.3(5)	8.9(4)
	1200	3.8(5)	1.2(4)	2.6(5)	1.4(5)
	1800	5.2(4)	1.0(3)	5.4(4)	3.6(4)
	2400	1.3(3)	-	2.9(4)	3.0(4)
OH at 3 km (molecules/cm^3)	0700	4.4(3)	8.1(2)	3.3(2)	2.6(2)
	1200	9.0(4)	5.8(3)	5.2(3)	7.3(3)
	1800	3.9(4)	4.0(3)	5.1(3)	1.2(4)
	2400	5.5(3)	-	8.3(2)	5.5(2)

[a] Cases are defined as follows: 1, without heterogeneous removal, no clouds, with stratospheric sources and surface source excluding HCs; 2, heterogeneous removal included; 3, HC sources included; and 4, cloud at level 6 included.

Table 5 shows predicted O_3 and OH values at selected times and elevations for the following cases:

- Without heterogeneous removal, with stratospheric sources, surface sources excluding HC sources and no clouds;
- With heterogeneous removal, with stratospheric sources, surface sources excluding HC sources and no clouds;
- With heterogeneous removal, with stratospheric sources, surface sources including HC sources and no clouds; and
- With heterogeneous removal, with stratospheric sources, surface sources including HC sources and clouds.

As shown, the O_3 and OH concentrations decrease when heterogeneous removal is included in the model (see runs 1 and 2). Without heterogeneous removal, there is a higher concentration of the soluble species (e.g., H_2O_2, HNO_3, HCHO, PAN, etc.) and higher free radical concentrations (e.g., OH, HO_2, RO_2, etc.). In addition, the NO concentration is lower and the NO_2 concentration is higher without heterogeneous removal. Furthermore, the O_3 and OH concentrations increase when organic surface sources are included (see runs 2 and 3).

When clouds are included, the O_3 and OH concentrations decrease below the cloud due to a decrease in solar actinic flux in this region. The OH concentration immediately above the cloud increases due to backscattering (see runs 3 and 4).

Ozone concentrations predicted over a 48-h period for the conditions corresponding to case 4 in Table 5 constant surface sources) are shown in Figure 6. In the lower troposphere (up to approximately 2 km), a diurnal

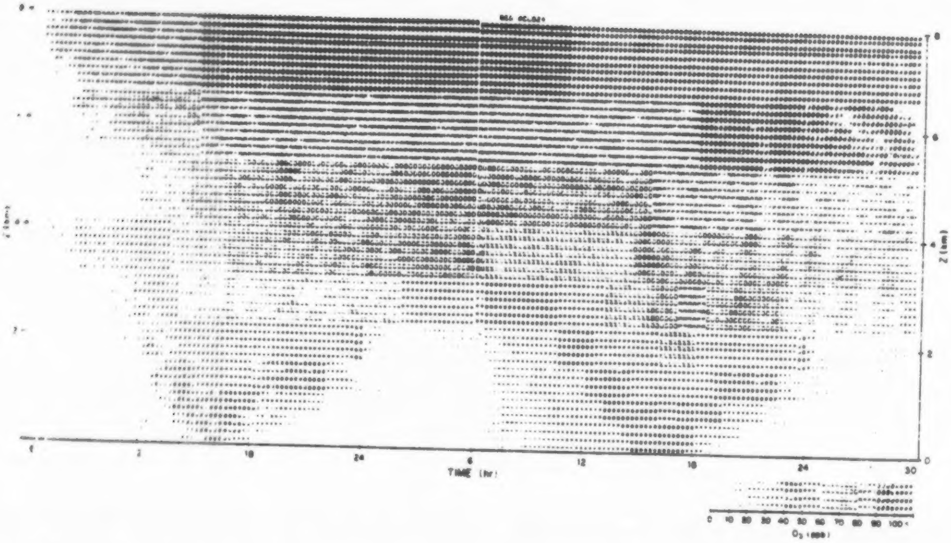
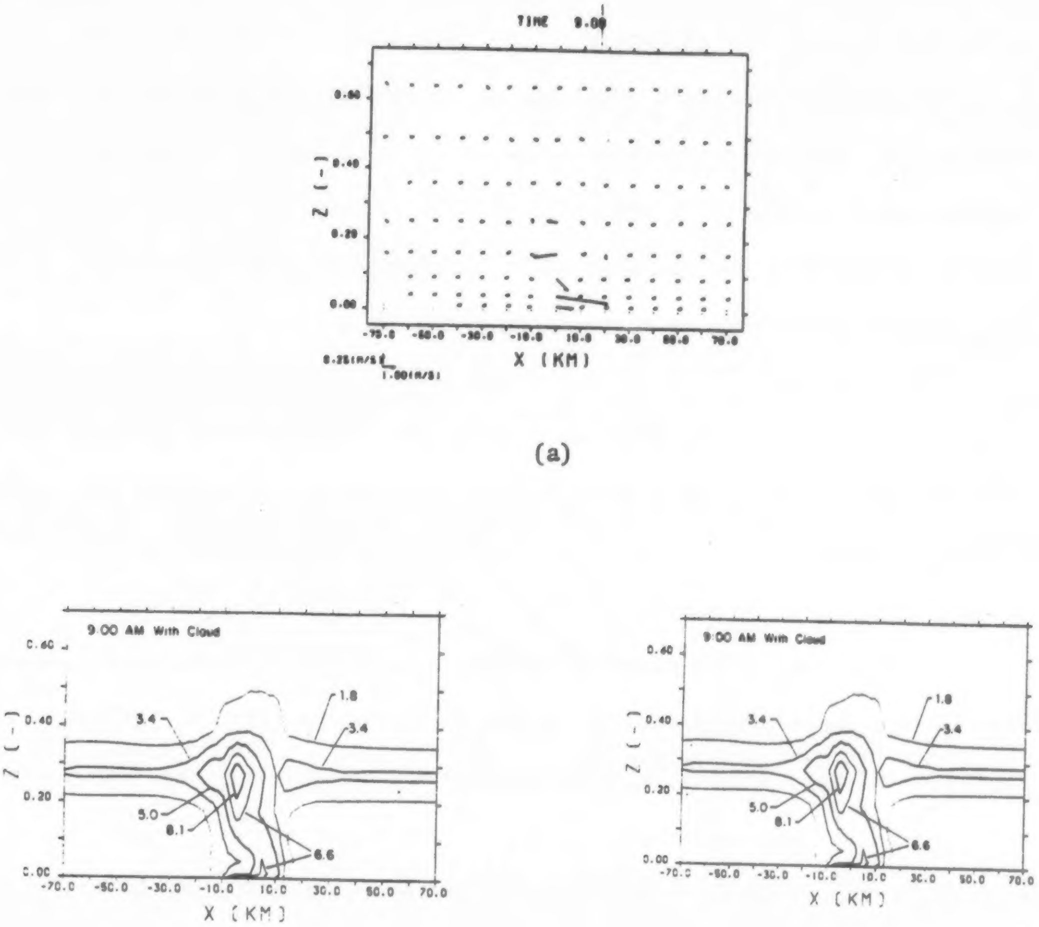


Figure 6. Predicted O_3 concentrations over a 48-h period for conditions given for Case 4 in Table 5.

profile is depicted, in which the O_3 concentration reaches maximum level in the late afternoon and minimum at night. Surface O_3 concentrations gradually fall to near zero in the early evening and increase sharply at sunrise.

A two-dimensional version of the STEM model for photochemicals is being applied to a coastal region in Japan. Figure 7 shows sulfate profiles calculated by using flow fields derived from a land/sea breeze model (Kitada et al., 1983). The land/sea interface is located at X= -5 km (X < - 5 km represents the sea region), and initial profiles of the primary pollutants representative of the land and sea regions were assigned. Shown are the wind field (Figure 7) and the concentration profiles at 9:00 a.m. for conditions with and without clouds (clouds located at z = 0.25). The without-cloud contours



(a)

(b)

(c)

Figure 7. Predicted sulfate concentrations for a land/sea breeze situation. Initial conditions were specified at 6:00 a.m. Concentrations are x 10^2 ppb. (a) wind field, (b) concentration profile without clouds, (c) concentration profile with clouds.

show the circulation patterns at 9:00 a.m. A strong surface inland flow is predicted at the land/sea interface, with a strong circulation cell extending to $z \simeq 0.25$. The resulting sulfate contours depict this general circulation. In the absence of clouds, the sulfate peak is located about 10 km inland and $z \simeq 0.1$. In the presence of clouds, the peak is located at the land/sea interface at cloud level. The sulfate levels are lower in the below-cloud region due to the reduced solar actinic flux and the lower SO_2 concentrations resulting from the removal of SO_2 from the air mass as it circulates through the clouds. In-cloud sulfate production is shown along $z = 0.25$.

The computation time for this two-dimensional code with 330 grid cells, that is, $(v,z) = (30,11)$, for a 24-h simulation is approximately 940 s on a FACOM M200 computer.

Three-dimensional simulations using the entire model have been successfully performed for a short period over a limited region on an IBM-370. However, we are currently using a simplified three-dimensional version for two transport species and 27 chemical reactions) (actually the first-generation STEM) to study SO_2/sulfate transport in the eastern United States (Carmichael and Peters, 1983). Figure 8 displays predicted 24-h averaged SO_2 and sulfate concentrations calculated with July 4, 1974, meteorological data. Surface concentrations and vertical profiles along the indicated slices are displayed. The CPU computation time for 9,504 grid cells, that is, $(x,y,z) = (27,32,11)$, is approximately 6 s per time step, or $\simeq$ 576 s/24-h simulation on the NASA-Langley Cyber 203 computer.

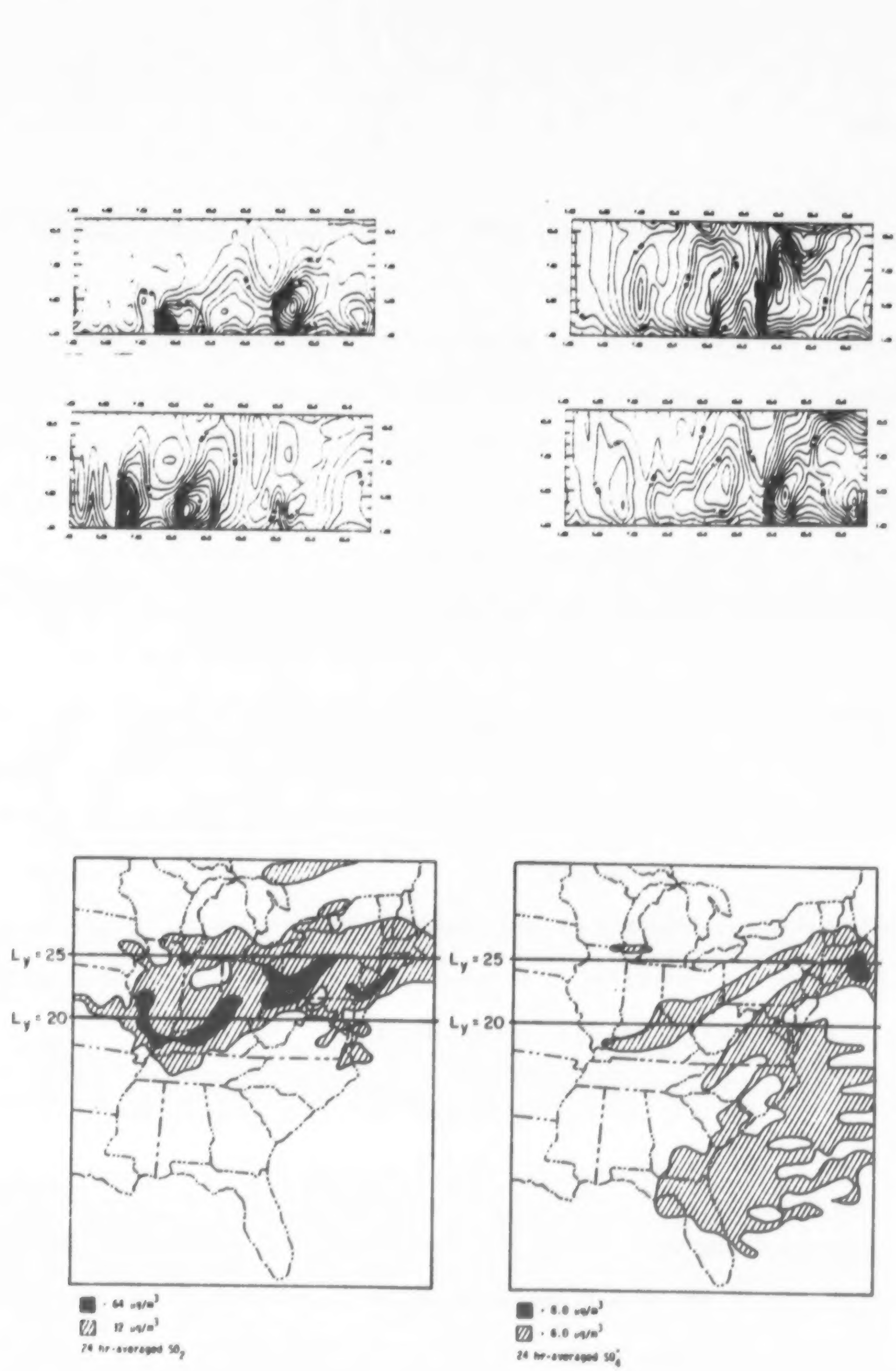

$L_y = 25$
$L_y = 20$
$L_y = 25$
$L_y = 20$
■ - 64 μg/m³
▨ 32 μg/m³
24 hr-averaged SO_2
■ - 8.0 μg/m³
▨ - 6.0 μg/m³
24 hr-averaged $SO_4^=$

Figure 8. Predicted 24-h averaged SO_2 and sulfate concentrations for conditions on July 4, 1974. Top figures and Z-X slices correspond to Ly = 20 (top) and 25 (bottom).

CONCLUSIONS

A combined transport/chemistry model for the regional-scale transport of photochemicals and their precursors has been described. The model, which is Eulerian and three-dimensional, is an extension of an operational SO_x transport model developed by the authors. This second-generation model treats 19 species as transported species, including NO, NO_2, SO_2, $SO_4^=$, O_3, HNO_3, NH_3, PAN, and HC. Both heterogeneous and homogeneous chemical reactions are modeled, as are both wet and dry removal processes. The model employs a finite element numerical method and performs the nonlinear chemistry with a pseudo-linearization scheme, resulting in virtually decoupled numerical calculations.

The model is sufficiently detailed to simulate urban boundary layers as well as non-urban-free troposphere chemistry, boundary-layer-free troposphere exchange in cloud-free and cloudy environments, and in-cloud and below-cloud wet removal and chemistry processes. The model is capable of addressing leading regional-scale scientific as well as regulatory problems.

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DISCUSSION

J. Novak: You mentioned a lot about cloud parameterization and the temperature in cloud water. How did you get that data?

G. Carmichael: We are currently testing this model and, if it is available, that is the parameter. There are two directions to go with our plans. The first is to incorporate a cloud-type model, say a pluvial-type model, in which you can generate cloud from surface parameters and the rainfall grade. And from the lookup table if Uranius model (not the transport chemistry model but the cloud variation model for a number of cases), you could then, instead of running a model, have a lookup model approach. From those parameters, you would get the statistics for that storm or that cloud.

Maybe the third approach is to couple them with a dynamic model where you would be predicting and to tag this along with the dynamic model. There is no easy way of doing it.

B. Luebkert: Is your model based on the principle that you have different submodels like the Lamb model such that you can basically interchange modules, like the chemistry or the cloud module?

G. Carmichael: Yes, this model is modular, so you can interchange these processes.

J. Killus: Was I correct in reading the slide you had for surface deposition rates for O_3, that there is something like 50 times less than that residue?

G. Carmichael: Ozone is--I cannot remember the numbers.

J. Killus: It looked like 0.01 and 0.1.

G. Carmichael: Yes, it was low for O_3.

J. Killus: That seems to conflict with practically every bit of data that I am aware of for O_3 and NO_2 surface deposition rates. Those have actually been measured in closed chambers and so forth, and they were always comparable to SO_2. Was this based on any existing data?

G. Carmichael: This was the procedure that we used, based on calculating the surface resistance for a species that we know a lot about like SO_2 and then charaterizing and modifying that surface resistance--according to the chemical-physical nature of the module. However, that is not hot-wired into the program.

G. Whitten: Earlier, you illustrated the dramatic effect of NO_3 with cloud cover. Later, you illustrated the dramatic effect of cloud cover on nitric acid, but you used the expression "NO_3." Do you mean the nitrate radical or the nitrate ion?

G. Carmichael: Nitrate ion.

G. Whitten: In all cases?

G. Carmichael: No, the earlier slide was of the gas phase of NO_3.

G. Whitten: The NO_3 gas-phase radical?

G. Carmichael: Yes.

ACID DEPOSITION AND OXIDANT MODEL*

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MODEL DESCRIPTION

The Acid Deposition and Oxidant Model (ADOM) is used primarily to simulate acid deposition. However, oxidant transport is also simulated, since oxidants are products of the chemistry model.

The issue of nonlinearity of the transformation of SO_2 to SO_4^{2-} in the atmosphere is important when one wishes to quantify long-distance, source-receptor relationships. Modeling efforts on the long-range transport of acidic pollutants have today been confined to the linear parameterization of the transformation processes, due in part to the lack of data and the inadequate modeling techniques of these processes.

Over the past years, the quantity and quality of atmospheric chemistry data have improved significantly. Also, atmospheric chemical transformation modeling techniques have shown improvements and promise. It would therefore be desirable to develop a long-range transport model that includes nonlinear chemical reactions in the atmosphere, with the aim of studying the effects of the nonlinear processes on the source-receptor relationship at distances greater than 100 km. The model would be designed for regulatory applications.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Three organizations, the Ontario Ministry of the Environment, Environment Canada, and the Federal Republic of Germany are jointly funding the development of such a model. The spatial domains of interest are different for the three sponsors. The attached map, a polar stereographic projection true at 60° N latitude and used by CMC for numerical weather forecasting, delineates the boundaries of the domains of interest to the Ontario Ministry of the Environment (inner domain) and Environment Canada (outer domain). The grid size (X,Y) is 127 km x 127 km. The Federal Republic of Germany is interested in a geographical domain in Europe and has a somewhat lesser grid size.

The pollutants of interest are SO_x, NO_x, and O_3. The model has a time resolution of 1 h and is designed to simulate pollutant concentrations and deposition over the domains of interest for episodes lasting from 24 to 96 h.

The problems to be addressed by the model are: the source-receptor relationships for SO_4^{2-}, NO_{3-} depositions for a travel distance of 1,000 km, and the formation and transport of oxidants in the same space and time scales. Because the model is Eulerian by design, these problems can easily be addressed by executing the model with appropriate emissions inventory and input data in the given domains. Although the model is aimed at episode simulations, long-term averages can be simulated either by averaging over a finite number of typical episodes or by executing a simpler version of the model for a period of 1 yr.

The model is being developed by Environmental Research and Technology, Inc. (Concord, Massachusetts, U.S.A.) and Meteorological and Environmental Planning Ltd. (Downsview, Ontario, Canada). A scientific review panel consisting of 12 internationally reputed scientists is overseeing the model development. Development of the model should be complete by March 31, 1986. A 1-yr time period will then be required to evaluate model performance.

The joint development of the model by the Ontario Ministry of the Environment, Environment Canada, and the Federal Republic of Germany is based on a Memorandum of Understanding mutually signed by these agencies. Cooperation from other interested agencies is welcomed and can be arranged through similar institutional agreements.

Difficulties in achieving the established goals are expected to be primarily of a technical nature, particularly in obtaining adequate data to specify the flux fields at the required spatial and temporal resolution and to give useful evaluation data sets. Specifically, we can identify potential problems in the specification of:

- Mass consistent wind, humidity, cloud, precipitation, and radiation fields with a 6- to 12-layer vertical resolution and horizontal grid size of 150 km or less appropriate for oxidant modeling.
- Emissions inventories of NO_x and speciated HCs together with concentration distributions (in three dimensions) of appropriate gas and aerosol-reacting consitituents required for the complex nonlinear chemistry models.

This will surely introduce uncertainties to model results; e.g., one may have to specify typical values to execute the model. The availability and quality of such data will likely improve in the future, making it possible for the model, in view of its modular nature, to perform better. In fact, model runs may provide both incentive and information on the nature of the improved data sets required. This should perhaps be a major item for discussion at this conference.

ACKNOWLEDGMENTS

This model is jointly funded by the Ontario Ministry of the Environment, Environment Canada, and the Federal Republic of Germany.

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DISCUSSION

B. Luebkert: Do you have any particular area in mind for validating this model and what plans are there to gather the emissions inventory?
P. Misra: We plan to develop the model for three special studies that were done in the past 2 or 3 yr. The studies were done in order to create an emissions inventory for that period and to collect data for that period.

B. Luebkert: Do you have all the emissions inventories you need, all of the HCs for example?

P. Misra: The development has already started and that includes comparison of emissions data for the model. That is going on at this time. So, we will see what is available.

E. Runca: Is the grid size of the model, the total grid size, 110 km?

P. Misra: That is right.

E. Runca: Is this a size that is enforced by the size of the region in which the model is to be applied?

P. Misra: That went into the selection of the model grid size.

E. Runca: If you want to do exposures and factors like cloud formation with the model. could the grid size be less than it is now?

P. Misra: It could, but we have not found any smaller grid size at this time.

F. Smith: Would you say a little about the horizontal diffusion? Does this really represent the mixing or is it really just for deformation?

P. Misra: That is right.

R. Yamartino: The deformation basically represents the differences in the horizontal wind field across the grid and to some extent the unresolved wind field.

F. Smith: Strictly speaking, a deformation is not a diffusion process. You are just squashing a lump of material and expanding it; it is a smashing of area.

R. Yamartino: Right, but we are essentially representing the advection by an mean flow. The rest of it at the smaller scales you choose to represent is incorporated into the diffusion process to allow for diffusion.

A. Venkatram: The deformation is nothing but the velocity gradient. The unresolved velocity is the velocity gradient multiplied by some land scale. It was used by Morenski in his numerical weather-prediction model. He has just taken it and put it into looking at the diffusion of the concentrations. In fact, the alternative way to do it is Bob Lamb's way, to run these latest realizations with ensembles and then look at the diffusion.

F. Smith: Also empirical diffusion.

A. Venkatram: This is empirical.

F Smith: Actually, it is a diffusion process. You say you actually work out a x,y and plug it into a diffusion equation somehow.

R. Lamb: I think the intent of it, as in this Morenski model, is that the horizontal shear is supposed to somehow generate small-scale fluctuations and a sort of variance velocity based on what the model-scale horizontal shear is doing. We come up with an estimate of a velocity variance, in other words, as that horizontal shear is beginning to break down into some sort of mesoscale fluctuation.

II. Eulerian model

(1) Adjustable but planned as 1 h.

(2) Adjustable but planned as 127 km on polar stereographc grid (true at 60°N).

(3) Two applications planned: 33 km x 33 km and 65 km x 65 km (i.e., approximately 3,500 and 7,000 km, respectively).

(4) Presently operational as 1-layer model but 12-layer and 20-layer versions are planned. Variable vertical resolution, approximately logarithmic.

(5) 5 km and 10 km, respectively.

(6) From a wind field model that uses PBL theory.

(7) Eulerian splines are interpolated to compute fluxes, which are then altered to avoid negative concentrations. Finite element scheme with filtering also available. Both schemes conserve mass, have low distortion, low phase error, and good peak preservation properties.

(8) Yes, predicted from w-equation.

(9) Transport as in (7). Diffusion from fully-implicit finite element algorithm.

(10) Proportional to velocity deformation tensor according to Smagorinsky.

(11) Kz obtained from similarity and convective scaling for convective conditions and from Brost and Wyngaard (1978) for stable conditions.

(12)

(a) Mixed depth variable in space and time.
(b) Sensible heat flux used to compute convective depth according to Maul (1980). Empirical relation z_i equals approximately 2,400 $u_*^{3/2}$ of Venkatram (1980) used for stable depth.
(c) Via the K_z profile.

(13) All cloud types to be treated but not in version to date.

(14) Model uses terrain following coordinates.

(15) Resistance modeling approach used so that deposition velocity depends on local surface roughness, canopy resistance, wind speed, and stability.

(16) Both rainout and washout effects to be computed from cloud module and heterogeneous chemistry module. Removal rates will be functions of space, time, and other variables.

(17) 7 s/grid point in the present 1-layer model.

(18) Estimated 1.7 x 10^6 bytes assuming 15 species and 1-layer but including storage of wet and dry surface fluxes and flux through top.

(19) Preliminary tests but on unfinished model.

(20) Full-scale tests expected in 1-2 yr.

III. Chemistry

(1) The chemical mechanism is interchangeable. The current chemical mechanism consists of four parts.

(a) Anthropogenic RHC/NO_x/SO_x based on Atkinson et al., 1982. Biogenic RHC/SO_x based on Lloyd et al., 1983. $SO_4^=$/NO_3^-/NH_4^+ aerosol composition based on Stelson and Seinfeld, 1982. Aqueous mechanism based on ERT, 1982. See references for species list.

(b) Concentrations of H_2O vapor and CO_2 are prescribed.

(c) Atkinson et al., 1982, Carbon II and III and Dodge, 1977, have been tested in urban scale simulations.

(d) See references for species list.

(e) H_2O vapor and CO_2.

(f) The number of species is user selected.

(g) Four- to seven-day runs are anticipated.

(2)

(a) The pseudo-steady-state approximation is used for very fast reacting species such as OH.

(b) Nighttime chemistry is included in the mechanisms. See references.

(c) Nighttime windshear, stability stratification and turbulence are accounted for through the 12-layer windfield and K-theory diffusion. The winds in layers between 850 and 500 mbar are derived from the numerical weather model and the lower wind from a numerical boundary layer model.

(3)

(a) Horizontal grid resolution is variable, with finer grid resolution in source areas which helps resolve subgrid concentrations. Explicit parameterization beyond this have not yet been developed.

(b) The key subrid scale chemical process of concern is fast reactions such as $NO + O_3 \rightarrow NO_2 + O_2$.

(4) Major point source emissions are injected into the layer of their final plume rise, based on the hourly meteorological condition. They are currently pseudo-diffused horizontally into the injection layer.

(5) The model does not provide a measure of how predicted grid average concentrations may vary from point observations. Sensitivity analysis with fine horizontal resolution is being performed to address this question.

(6)

(a) Photolytic rate constants vary with latitude, longitude, time of day, elevation, and cloud cover.

(b) Cumulus and frontal clouds, vertical velocities, and entrainment rates are obtained from a one-dimensional cloud model and are applied to the fraction of the grid column covered by clouds.

(7) Natural sources of HCs and NO_x are simulated explicitly by input of emissions and chemistry based on isoprene and α-pinene oxidation. Stratospheric O_3 and NO_x can be entrained from aloft.

The NATO/CCMS AIR POLLUTION MODEL COMPARISON*

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INTRODUCTION

In 1980, the North Atlantic Treaty Organization/Committee on the Challenges of a Modern Society (NATO/CCMS) initiated a pilot study on air pollution control strategies and impact modeling. The study was undertaken by three panels covering three separate areas--emissions, air quality prediction, and environmental impact.

The air quality prediction panel, which was chaired by representatives from The Netherlands, was charged with producing documents describing and comparing models for interregional transport of air pollutants, with an emphasis on the transport, removal, and transformation processes of SO_2 and NO_x. The results will be reported in four separate documents covering the following topics:

- state of the art of interregional modeling,
- comparison of interregional models,
- removal and transformation processes, and
- interpretation and summary of documents 1 through 3.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Documents 1 and 3 have now been completed and are available as NATO/CCMS Reports 126 and 127, respectively. Documents 2 and 4 are in preparation.

This paper briefly reports the main results and progress of the panel to date.

STATE OF THE ART OF INTERREGIONAL MODELING

This document (NATO/CCMS Report 126) reviews operational models that describe dispersion, deposition, and chemical transformation processes on a spatial scale ranging from 50 to 5,000 km. This range is subdivided into the mesoscale (50 to 500 km) and the synoptic scale (500 to 5,000 km). Depending on the spatial scale, different approaches to observations and physical/mathematical descriptions for the models are required, as indicated in Table 1.

All transport models are based on the mass conservation equation and use either a Lagrangian or an Eulerian framework. The Lagrangian approach, in which any desired polluted air parcel is followed (or traced) along its path through the atmosphere, is much simpler to consider from a numerical point of view. It has, however, certain disadvantages when applied to multiple source areas or (complex) chemistry. Eulerian models require a medium- or large-sized computer and considerable computer time, and thus incur high development and operationalcosts. Moreover, their mathematical complexity hinders fast implementation in operational air pollution abatement programs.

TABLE 1. A CLASSIFICATION OF MODELS ACCORDING TO SPATIAL SCALES

Model Type	Range (km)	Total Model Configuration			Data Frequency (measurements/h)
		Observation Used	Wind Field	Diffusion	
Microscale	0 to 50	surface synoptic data or special measurements	uniform or varying in time and space	homogeneous (dispersion coefficients or homogeneous eddy diffusivity concept)	~1
Mesoscale	50 to 500	surface synoptic data and radiosonde data	varying in time and space	nonhomogeneous eddy diffusivity concept)	1 - 0.3
Synoptic scale	50 to 5,000	data and radiosonde	varying in time and space	nonhomogeneous (eddy diffusivity concept) or instantaneous mixing	0.3 - 0.1

While suspended in the atmosphere, emitted matter is exposed to photochemistry and the removal processes, dry and wet deposition. The direct, adverse effects of high air pollution levels (e.g., on human health) have been reduced considerably in urbanized, industrial areas via site planning and high-stack policies. It has become increasingly clear that the deposition process is the major environmental impact of air pollution. No operational models exist, partly because a quantitative description of dry and wet deposition requires a detailed knowledge of both atmospheric dynamics and chemistry.

REMOVAL AND TRANSFORMATION PROCESSES

The document on removal and transformation processes (NATO/CCMS Report 127) focuses on SO_x and NO_x in the atmosphere.

Dry deposition can be satisfactorily parameterized by using the deposition velocity concept, and a wealth of definitive data already exists on dry deposition for various pollutants and surfaces. Although similar concepts exist for wet deposition, the underlying physical and chemical processes that lead to the wet deposition of matter are neither fully understood nor fully validated.

For reasons of simplicity, chemical conversion is often described by linear decay. Current experimental evidence allows an estimation of the constant of proportionality.

The underlying series of chemical processes is extremely complicated; the most relevant chemical processes involving NO_x consist of more than 20 reactions. The study of these reactions is a separate discipline still far from completion. Because of limitations imposed by current computer capacity, the transport and chemistry of no more than one or two dozen air pollution components can be described satisfactorily on an operational basis.

COMPARISON OF INTERREGIONAL TRANSPORT MODELS

This document, which is now in preparation, is restricted to a comparison of models that operate on a mesoscale not exceeding 500 km. The models evaluated meet the following criteria, which were determined by the panel of investigators:

- The model output consists of ambient concentrations and deposition (dry and wet) estimates;
- The time resolution is less than 3 h;
- The horizontal spatial resolution is 10 to 25 km;
- The vertical atmospheric structure is resolved (i.e., no single-layer models);
- Advection, turbulent diffusion and deposition (dry and wet), and linear chemistry is incorporated;
- A complex source area is described; and
- Only routinely available meteorological input data are used.

Four models were selected for comparison: (1) the TDMB model (Klug et al., FRG); (2) the SAI model, Dutch version (Reynolds and Builtjes, The Netherlands);

(3) the RIV model (Van Egmond et al., Institute for Public Health, The Netherlands); and (4) the KNMI model (Van Dop et al., The Netherlands).

The comparison was conducted in Darmstadt, FRG, under the coordination of Klug, and supported by NATO/CCMS. The Netherlands and surrounding areas were selected as the test area because:

- The region contains a dense network of meteorological surfaces and radiosonde stations;
- SO_2, NO_x, and O_3 are routinely monitored bv networks in the involved countries, and data can thus be easily made available; and
- A detailed emissions inventory for the area exists.

The comparison consisted of three phases. In Phase 1, the elementary numerical procedures in all models were compared. For this purpose, the dispersion of a single point source under uniform meteorological conditions was simulated. In Phase 2, the dispersion of a single source under real meteorological conditions was simulated. For this purpose, three 48-h episodes with varying meteorological conditions were used. In Phase 3, the standard emissions inventory was input to each model and runs were made for the three selected episodes. A sensitivity analysis was performed, and the differences in model outcome were compared.

PRELIMINARY RESULTS AND CONCLUSIONS

The study, which is now in the final stage, will be reported towards the end of 1983. Some preliminary results and conclusions are as follows:

- Schemes should be chosen with care. Saving computer time by increasing grid size or simplifying numerical procedures leads to unacceptable numerical errors.
- Ideally, inflow conditions at model boundaries should be specified. However, the lack of data prevents this in most cases. The best alternative is to choose boundaries in regions where no high concentrations of involved compounds are expected and to assume zero inflow. In all other cases, artificial boundary conditions will lead to errors, which are often difficult to estimate.
- The rate of vertical turbulent diffusion (and thus the height to which released matter may extend) is of paramount importance in determining the concentration of a single pollutant. For operational applications, this diffusion rate should be estimated from routinely available weather data. Although some progress has been made in this area, it remains a matter of concern.

GENERAL DISCUSSION FOLLOWING SESSION I

Jack Shreffler, Chairman

J. Shreffler: Does anyone have questions for the people who presented papers today or questions for the OECD people?

E. Runca: I would like to discuss a concept that was brought to the attention of the audience several times today. That is the concept of a simplified, sophisticated model.

This meeting is addressing the long-range transport of air pollutants and the formation of oxidants. I think that priority should at least be given to the processes that have to be considered in order to describe at least what is happening and what we can assimilate. Then, we come to the issue of application and the parameterization of these processes in order to reach a temporary solution and to reach a greater solution.

I think it is more appropriate to look at what we want to describe and to identify the elemental processes. After we have identified these processes, we can consider how we can eventually simplify them in such a way to "steer" the model with at least a description of the processes you want to assimilate.

D. Jost: I agree with Mr. Runca's more philosophical point. However, in preparing for this workshop, our U.S. colleagues sent questionnaires to the people who were presenting models requesting that they follow some scheme to describe which processes are handled in their model. Additionally, questions were prepared concerning the objectives of this workshop.

Thus, I am asking the participants, especially the chairmen for the small group discussions to occur tomorrow, to keep in mind Mr. Runca's comments, the questionnaire that has been prepared, and the objectives that have been mentioned.

S. Zwerver: One point is not clear to me. We will be discussing these models by starting with the objectives and looking at how capable the models are of meeting these objectives.

However, there is another way. The objectives can be discussed as possibilities for models. Can the modelers say something about how changing the objectives would influence the simplicity of their models? Perhaps that is impossible, but I think the politicians or policymakers in the different countries are not completely aware of the detailed objectives they have. It would be helpful to determine the exact definitions of the objectives and to look how these objectives are obtained and what it means so that we can find the most optimum point.

A. Venkatram: A lot models have been described today, ranging from the basically simple to the overwhelmingly complex model. It occurs to me that the reason for complexity is really understanding, more specifically the quality of understanding, because we can never really hope to quantitatively simulate the atmosphere, because we can never hope to have the input. Thus, it seems to me that the simple models are obviously missing something. That is the implication of complexity. I would like for the complex modelers to identify those missing pieces in the simple models.

A. Venkatram: Why should we make models complex, unless the simple models are missing something--missing something qualitatively?

E. Runca: That is exactly my point. The things we have seen in complex models must identify with the processes that are relevant in relation to modeling regional air pollution problems.

My interpretation of these comments is that, by discussing the simple models with the authors, we can have an understanding again of the the processes to be taken into account when modeling long-range transport.

J. Shreffler: That philosophy drives you to a complex model with high resource requirements and high data requirements. As I understand it, part of the purpose of this meeting is to make recommendations to OECD on the way they should go. Certainly the resource requirements are part of it.

A. Galli: We are getting bogged down with the modelers issue and that is just part of what might be a bigger problem with emissions inventory data bases. Before we can answer the questions just presented, we need to get the European community, Canada, and the U.S. to upgrade their emissions inventories and their data bases, in order to build a better model, a simpler model or a more complex model, and then have another data base in order to test them.

So, it might be premature to even discuss here the simple model versus the complex model. The first thing we have to discuss here are data bases, the availability of data bases and how good they are. Then we can consider how simple or how complex the models should be. A big argument is taking place in this country as to whether building more complex models buys you anything, as you are inferring here, versus spending the money on emissions inventories and data bases. That is not going to make the modelers happy; nonetheless, it has to be recognized, because the models are only as good as the input data.

A. Christie: There is no question about that, but the emissions inventories are no good unless you know what is going to happen to them once you have admitted them. Surely a part of our consideration should be the state of emissions inventories and concentration measurements at the present time, and what is likely to be required by some of the more complex models that are on line and are likely to require data bases. We really should not be approaching this from the point of view of designing our emissions inventories to what we think we can produce and then bending the models to fit this, because we may lose something in the process.

A. Galli: You already have a number of models. Those models tell you that you need some kind of input data, whether they are emissions inventories or aerometric data. You really do not have enough good, scientifically valid data, with which to build models and later test them throughout Europe and North America, regardless of whether they are simple models or not.

I am not suggesting that the emissions inventories drive the models. I am saying that you have to have a number of models now that require certain types of information.

A. Christie: We are talking about how one validates a model in its fullest sense. In discussing his model, Gregory Carmichael showed the aqueous-phase process. However, these models are based on a knowledge of droplet size distribution and a variety of parameters that may or may not be known.

How can we parameterize the laboratory-scale model that puts this into it until we at least have some measurements that can validate even a one-dimensional or two-dimensional model on a smaller scale? We have to agree on what you have to have parameterized to be able to put anything into the models on the scale most of us are looking at, that is, 100 km.

J. Killus: Are we certain that we know what we are talking about when we say complex models versus simple models? For example, we heard today that the Hov model or the U.K. model were certainly meteorologically simpler than any of the grid models being discussed. On the other hand, the chemistry they are using is extraordinarily complex, vastly more so than you could possibly include in any sort of grid formulation.

In the urban modeling problem and in the regional modeling problem, we have found that transport, in developing a wind field and mixing layers, is the critical factor where one can get away with fairly simple biochemistry. However, that may not always be true. In certain circumstances, like the stratosphere, it is not true because the chemistry must become quite complex to deal with the situation.

I would really like for us to solve for model application in conjunction with development. Only when you apply the model, only when you are looking for real data and understanding exactly what sort of situation you are dealing with do you decide the appropriate scale and complexity required for the model.

J. Shreffler: That is a very good point. Only four, five, or six models were presented, so that probably sums up the efforts on the regional scale. None of them is a simple model.

D. Jost: Within OECD, all this business started about 15 yr ago with long-range transport and acid deposition. Within this field, we knew which pollutant we were looking at, which was mainly SO_2. All this business has been done during the last year, mainly for SO_2 and as it occurs.

In the field of oxidants, I get the feeling that we are not even very sure which pollutants are the important ones or which pollutant needs to be abated.

During this meeting several chemistries have been presented, more chemistries even than models. Do the people who are using these different chemistries know if they all give the same results or if they are different? Do the chemistries all indicate that we need to abate primarily HCs instead of NO_x, or are there also applied chemistries that give comparable results?

R. van Aalst: We have been trying to compare different chemical models and, indeed, they tend to give different answers. To my knowledge, several examples of such differences have been reported in the literature.

Most of the problem is identifying the kinds of concentration regimes these differences show. I agree that putting in a complex chemistry will lead you in a specific consideration to get rid of some difficult intermediate reaction, which could be vital in other specific situations.

So, the policy probably is that you should get agreement upon the fundamental chemistry and decide in the actual situation you are modeling what you should leave out. Even with respect to the fundamental chemistry, there seems to be differences at the present between the chemical models in operation.

G. Whitten: The testing of chemical modules is something you can do outside of the atmosphere with the large data base that exists for smog chambers. We are now reaching the point where we can characterize the background effects within smog chambers. When we see different control strategy effects among different chemical mechanisms, we can analyze the specific reactions and parts of the chemistry that have lead to these effects, and we can design smog chamber effects to emphasize that part of the chemistry and then test them at that level.

EPA has been sponsoring this ongoing research for many years. Scientists at Systems Applications, Inc., as well as Dr. van Aalst have also been doing some of this research. Mainly, you have to get into a position where you can see the specific reactions to the different parts of the chemistry because you quite often have a very complex chemistry and many, many reactions. Yet, the particular reaction that is involved might be a very simple one and might not even be in the complex chemistry. So the complexity in the chemistry is not always realistic to the problem involved in the differences in control strategy and how they are used.

R. Lamb: Assuming for the moment that we have a rough definition of complex and simple, the answer to the question lies in what you are trying to predict. If you are interested in predicting hourly average O_3 at any place in a region, then you have to look at all the processes that are instrumental in affecting that level and then you have to determine what it is you needed in the way of data and parameterizations and the things needed to predict that. If you are interested in the annual average O_3 over all of Europe, then you can obviously get away with several models.

In my view, the answer to the question has to be posed by starting with what it is you are trying to get at and then by going back. You can also ask the question in this way: Given the data that we have, given the knowledge we have at this moment, what can we say?

What we can say may be quite limited. If it is limited, if we cannot make meaningful statements about the things we are interested in, then we have to determine what more we need to know and what it is going to cost to get that information?

So, the answer is quite simple. It starts with what you are trying to predict. If you want to predict an hourly average concentration and you insist on a simple model, the prediction of that model is going to be unreliable. Thus, we are trying to minimize that error by going to more complexity and so on. We really need a sophisticated model to determine the level of sophistication we need, because we cannot guess which of the processes is dominating and which of them is important. We almost have to have a complex model to answer that question itself. It seems to me that all roads lead back to a complex model.

B. Dimitriades: Obviously, you can get to the annual average by using the complex model and averaging the hourly data, or you can get to the same answer by usng a simple model, which gives you the annual average. The key question here is: How do you accomplish significantly more accuracy by choosing a simple model or a complex model? This is really relative by going the simpler route. That is the key question.

R. Lamb: If you write out the equation to predict annual average O_3 in the case you just raised, you find turns in the equation that you do not know how to resolve. So, you begin guessing at them. When you make a guess, you may be wrong and you may end up with a prediction that is unreliable.

If you grind through hour by hour, you get around to the assumption you made to go directly to this number. So, you are again back to a complex model. You do not know whether the guess you made about those terms is accurate.

This is the problem of closure. Some closure schemes work well for some problems, but fail for others. So a priori in these complex problems with nonlinear chemistry, diffusion, transport, all these interaction mechanisms, a guess at a closure scheme in advance is a very speculative thing.

S. Zwerver: The question is not to predict hourly averages of O_3 or annual averages of O_3. The question is: Will a 50% reduction of NO_x, an overall reduction, also result in a 10%, 15%, or 25% reduction of O_3. That is a different question? It could also lead to different ojectives for our models.

B. Dimitriades: A 50% reduction of what, O_3? The annual average?

S. Zwerver: I should say for the overall pattern of O_3, and that includes the probability of having high values in the overall areas.

A. Eliassen: After listening to this, I think that it would be useful in tomorrow's sessions to discuss the different models that we are trying to identify, what each can do, and where it can be applied. All of the models have their assumptions, and there are situations where they can say something useful and others where they cannot.

G. Whitten: Quite often in physics, the utilization of what is known as perturbation theory leads to a different philosophical approach. You have a mainline effect, and then you are aware of certain subtleties that would perturb that mainline effect.

These can be handled very accurately; it is not a matter of guessing.

Furthermore, there are many powerful mathematical treatments to handle such things that are small effects, once you have the main thing.

One of the risks that you run by going to a very large complex scheme and grinding away is that there is a certain sort of numerical diffusion. Over a long period of time, that can also add to a large error. Taking a very simplistic view and adding small perturbations tend to cut down on the accumulated numerical problem.

A. Christie: When you are addressing a prediction of the mean, you can certainly parameterize the area for which you want to predict the mean. If in fact you want to produce hourly values, you are not going to get that from a perturbation approach.

SESSION II

AVAILABLE EMISSIONS INVENTORY DATA BASES

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NORTHEAST CORRIDOR REGIONAL MODELING PROJECT EMISSIONS INVENTORY*

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INTRODUCTION

The U.S. Environmental Protection Agency (EPA), recognizing the potential impact of high ambient O_3 concentrations in the populous and industrialized Northeast United States, has initiated the Northeast Corridor Regional Modeling Project (NECRMP) in order to develop optimally effective and equitable strategies for O_3 control in the Northeast. The project includes several critical elements: (1) the development of comprehensive emissions, air quality, and meteorological data bases required for adequate technical analysis; (2) the development of a regional-scale photochemical oxidant model to consider interurban pollutant transport and photochemical transformation processes; and (3) the combined application of regional- and urban-scale models to evaluate alternative control stategies.

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*This paper has been reviewed by the Environmental Sciences Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

[†] On assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce.

The emissions inventory requirements for the EPA Regional Oxidant Model (ROM) are more detailed and extensive than those for urban models or control strategy analyses. Existing emissions inventories available to the modeling community are inadequate for regional model testing, refinement, and validation. Thus, EPA, in conjunction with the Northeast Corridor States, local agencies, and Metropolitan Planning Organizations (MPOs), has compiled an improved emissions inventory, which is believed to be a reasonably comprehensive and accurate 1979/1980 data base.

GCA Corporation's Technology Division was contracted to compile, provide quality assurance for, and correct the U.S. annual point and area source emissions inventory, including mobile sources. This has been accomplished through direct liason with the assistance from the state and local agencies. The Canadian emissions data were supplied by Environment Canada and the Ministry of the Environment.

BACKGROUND

The NECRMP emissions inventory study area includes all or parts of 14 states, the District of Columbia, and portions of Quebec and Ontario Provinces in Canada. The area lies between the boundaries of 69° to 82° west longitude and 38° to 45° north latitude, as shown in Figure 1. It includes in total Connecticut, Delaware, Maryland, Massachuetts, New Jersey, New York, Vermont, Pennsylvania, Rhode Island, and Washington, D.C., as well as portions of Maine, New Hampshire, Ohio, Virginia, West Virginia, and Quebec and Ontario, Canada. A substantial portion of the southeastern corner of this area

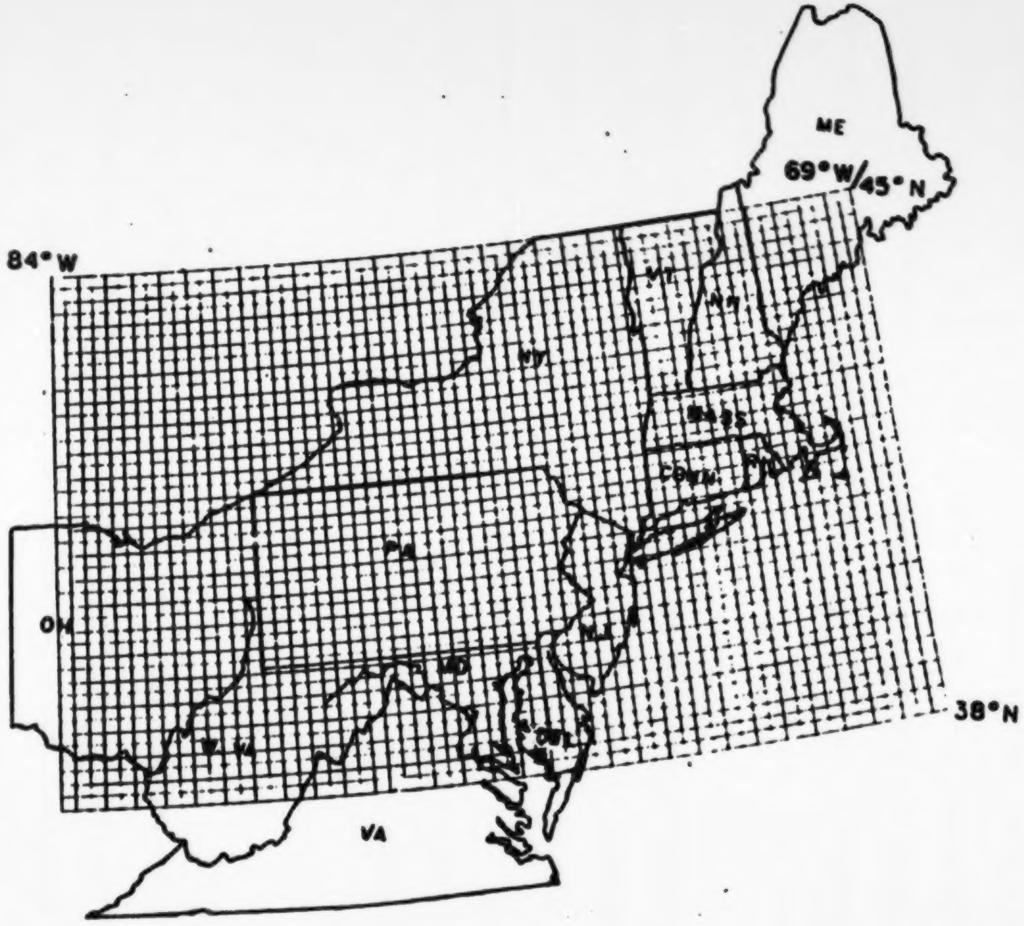
84°W
69°W/45°N
38°N
ME
VT
NH
NY
MASS
CONN
PA
OH
MD
W VA
VA
DEL

Figure 1. Boundaries of regional model grid system.

(about 175,000 km^2) is covered by the Atlantic Ocean. No emission estimates were made for this portion.

The data were allocated to a longitude/latitude system within the coordinates indicated above. A grid system of 1/6° latitude by 1/4° longitude was laid over the area and used as a basis for allocating area source emissions. The regional model domain extends 2° further west than the NECRMP emissions study area, to 84° west longitude. Thus, the standard regional model grid system consists of 2,520 grid cells (60 columns x 42 rows) of slightly varying area, approximately 18.5 km x 18.5 km. Emissions from the Ohio counties of Franklin, Licking, Perry, and Fairfield are the only values available within this extended 2° sector. Landsat data, population (census) data, and other similar information were also apportioned on the same basis for subsequent use in allocating emissions into these subcounty grids.

The major pollutants of interest were VOCs and NO_x, with CO included as a potentially important tracer. Particulate and SO_x emissions were also included, but no quality assurance or separate correction of source/emission information has been carried out for these pollutants, so their quality is undetermined. A distinction was made between VOC data supplied by states as "total" data and that supplied by states as "reactive" data, as the final modeler's file is required to be in a speciated format. A higher emphasis for completeness and accuracy of data was placed on point sources with VOC emissions above 500 tons/yr and NO_x emissions above 750 tons/yr, as they are generally found to be responsible for a large portion of the point source emissions and as they are likely to have distinct and isolatable impact on the model results.

The collection of raw emissions inventory data in the United States is generally the responsibility of individual states. The collection process includes sending questionnaires, filing permits, etc., and data are generally compiled via the state's own computerized data system or one supplied to the state and supported by EPA. EPA's state system is the Emissions Inventory System (EIS), a subsystem of the Comprehensive Data Handling System (CDHS). EIS is broken down in two major components, EIS/AS and EIS/PS, which handle area and point source data, respectiveley. After generating the raw data and the associated data files, states are required to annually supply the updated data to EPA's National Emissions Data System (NEDS), which is the national repository and data base upon which many national and state analyses are done.

Due to conflicts in resources, priorities, and differences in emphasis, not all states have kept their NEDS data files in a sufficiently detailed and up-to-date form for use in highly complex modeling exercises. Updates tend to reflect current program priority areas such as the requirement to update the data base for developing a revision to the State Implementation Plan (SIP), as required by the Clean Air Act. Hence, when it was determined that a data base of a more current and complete nature than available was required, it was necessary to identify the NECRMP effort as a state emphasis program. Consequently, EPA negotiated with each of the states in the study area to provide increased assistance and priority for updating the basic data base and for responding to the EPA contractor's questions and comments relating to compilation and quality assurance efforts. The basic NEDS/EIS formats and data were maintained as the starting point for continued improvements and error detection/correction. An additional emphasis to the effort for many jurisdictions included in NECRMP was the states' concurrent preparation of revised SIPs for O_3.

As indicated previously, states are normally required to update the NEDS data base annually. They may choose to complete updates more frequently, however. In the case of the NECRMP data, the data base compilation and quality assurance effort was planned as a one-time emphasis, and no further organized compilation thrusts are envisioned. However, emissions inventory is by nature dynamic; thus, further updates are to be expected form time to time to reflect detected errors or new/better information on sources and their emissions. Such updates would normally be improvements in estimates for the original base years of 1979/1980 as opposed to later "new" year data. As the program progresses, some separate activity may develop to project the 1979/1980 data base to a future year for strategy scenario for determining requirements to attain and maintain the National Ambient Air Quality Standards (NAAQS).

Legal constraints on emissions inventory data vary from state to state. EPA's authority under the Clean Air Act states that "emission data" shall not be deemed confidential, but determination of the legal meaning of emission data and its relevance to statutes on the protection of proprietary information are subject to complex, time-consuming procedures. During compilation of the NECRMP inventory, questions of confidentiality were not beyond reason. In cases where state statutes or state/industry agreements raised questions of confidentiality (and thus the release of data), EPA, the affected states, and the contractor were able to work out screens, selective deletions, etc., such that the data given to EPA and now in the NECRMP data base are not deemed confidential.

Specific data items that were claimed as confidential generally could be avoided or worked around in a manner that was not detrimental to the modeling aspects of the data collection program.

In Canada, the provincial agencies and regional offices are responsible for collecting emissions information that is primarily based on company-supplied data. Through surveys and questionnaires, other government agencies such as Statistics Canada; the Ministry of Industry, Trade and Commerce; and the Department of Energy, Mines, and Resources collect basic data that can be used in estimating emissions from major sources categories. There are no legal requirements for the submission of emissions data; however, many of the regulations controlling the release of toxic pollutants require quarterly or at least annual submissions of data that are used to develop emission estimates. The Canadian National Emissions Inventory is primarily used to assess the effectiveness of control programs. There are not formal restrictions on the release of company-supplied data, but informal agreements have been made regarding confidential processes. The Canadian inventory is updated every 2 yr. Background information on the Canadian emissions inventory was provided through a personal communication with Frank Vena, Air Pollution Control Directorate, Environmental Protection Service, Environment Canada, Quebec. Environment Canada has been extremely cooperative in supplying emission information for the NECRMP modeling efforts.

POINT SOURCE DATA

U.S. Inventory

A point source is typically defined as a stationary source large enough to be identified and tracked individually, usually emitting greater than 100 tons/yr of any pollutant of interest. However, all states in the Northeast Corridor did not strictly adhere to this specific definition. Each state submitted the most current point source inventory, typically for 1979/1980, in a computer-readable form compatible with either NEDS or the Emissions Inventory Subsystem/Permits and Registration (EIS/P&R), predecessor of the current EIS/PS and EIS/AS systems.

The following key parameters were among those reported:

- UTM coordinates,
- Stack parameters,
- Emission control equipment,
- Efficiency of primary and secondary control equipment,
- Operating schedule,
- Annual production rate,
- Sulfur and ash content of fuels, and
- Annual emissions by source classification code (SCC).

Stack parameters included height, diameter, temperature, and exhaust flow rate. Source location information, which was reported as UTM zone and coordinates to the nearest tenth of a kilometer, was converted to latitude and longitude in degrees, minutes, and seconds. Point source emissions data were reported for over 1,400 different SCCs. However, a large percentage of O_3 precursor emissions occurred within 10 to 20 categories. Table 1 lists the percentage of emissions from the major precursor-emitting point source categories. The categories listed account for 84% of the VOCs and 98% of the NO_x emitted from point soures in the NECRMP region.

Annual emissions from electric utilities were distributed seasonally by applying fuel and state-specific seasonal factors derived from U.S. Department of Energy (DOE) power generation statistics. Hourly allocation of electric utility emissions was based on hourly fuel use data collected from approximately 300 power plants during the Electric Power Research Institutes (EPRI) Sulfate Regional Experiment (SURE). Because of the lack of detailed data for developing specific temporal allocation factors for other point source categories, individual source operating pattern data were used to calculate temporal factors. Default values providing a uniform distribution were assumed when no operating data were available.

TABLE 1. MAJOR CATEGORIES OF OXIDANT PRECURSOR EMISSIONS IN THE NECRMP REGION

Category	Emissions	
	% NO_x	% VOC
Gasoline handling	<0.1	4.3
Petroleum refineries	0.8	7.4
Other chemical manufacture	0.2	3.5
Iron and steel manufacture	0.8	4.4
Stone, lay, glass, and concrete	1.1	1.0
In-process fuel use	0.2	12.9
Others industrial processes	0.5	18.0
Industrial surface coating	<0.1	17.2
Degreasing	0.6	2.0
Graphic arts	<0.1	4.8
External combustion boilers, electric generation	79.9	3.8
External combustion boilers, industrial	12.2	4.4
External combustion, space heaters - industrial	1.7	0.3

The following algorithms were used to perform the temporal allocation of annual point source emissions:

Standard Algorithm --

$$\text{Hourly Emission} = \text{Annual Emission} \times \text{Daily Fraction} \times \text{Hourly Fraction}$$

$$\text{Daily Fraction} = \frac{1}{\text{No. of Operating Weeks/Season} \times \text{Days}}$$

$$\text{Hourly Fraction} = \frac{1}{\text{Hours}}$$

Seasonally Weighted Operating Pattern Algorithm --

$$\text{Hourly Emission} = \text{Annual Emission} \times \text{Seasonal Factor} \times \frac{1}{13 \times \text{Days} \times \text{Hours}}$$

where seasonal factor = fraction of annual operation occurring in the season of study,

13 = assumed number of weeks of operation in a season,

days = number of operating days per week, and

hours = number of operating hours per day.

Uniform Default Algorithm --

$$\text{Hourly Emission} = \text{Annual Emission} \times \frac{1}{52\ \text{wk/yr} \times 5\ \text{days/wk} \times 8\ \text{h/day}}$$

where the hours of operation are assumed to begin in the eighth hour (0700-0800) local time.

The annual values reported by the states are estimates of actual pollutant emissions as emitted. The effects of control equipment have been incorporated by using type and efficiency information for primary and secondary control equipment. Approximately 40% of the emissions are estimated by using the NEDS standard emission factors with plant-specific information. About 25% of the emissions are calculated by using nonstandard emission factors and plant-specific data. Material balance (20%), source test (5%), and other methods (10%) account for the remaining reported emissions.

VOC Methodologies --

The chemistry mechanism in most photochemical grid models requires the lumping of individual VOC species into the reactive classes specifically treated by the mechanism. The current chemical mechanism in ROM handles four reactive classes: olefins, paraffins, aldehydes, and aromatics. Future mechanisms under development may require more classes or different distributions of individual chemical species in the designated classes. Thus, a general methodology was developed to calculate factors that, when applied to annual total HC or NMHC values, produce emissions for any reactive class defined to meet the requirements of the chosen chemical mechanism.

The basis of this flexibility is a set of 119 species profiles, each of which lists the typical organic compounds associated with a particular source category or process. An index file assigns each source category, represented by a unique SCC, to the most appropriate profile. Thus, given the definition of which organic compounds make up each reactive class, the mole fraction of each class in any profile is calculated. Mole factors for each profile and class combination are then determined by dividing the mole fraction for each class by the profile average molecular weight. These factors are then adjusted for aldehydes and for compatibility with HC that is reported as nonmethane or reactive VOC.

This generalized process, applicable for both point and area sources, enables the calculation of emissions for each reactive class of HCs required for any chemical mechanism a modeler may choose. These profiles also include specific information for splitting the NO and NO_2 components of NO_x for each applicable source category.

CANADIAN INVENTORY

The Canadian emissions data were submitted to EPA by Environment Canada in computer-compatible form. Point sources are considered to be major individual pollutant emitters. The 1976 point source data base contains the following key parameters: latitude, longitude, stack parameters, annual emissions by standard industrial classification (SIC), and seasonal variation of SO_x and NO_x. Stack parameters are height, diameter, exit temperature, and flow rate. The 1978 data base includes additional parameters such as base quantity, sulfur and ash content, and emission factor. Source location is reported in latitude/longitude by degrees, minutes, and seconds. Point source emissions data were reported for 62 different SICs. Table 2 list the major contributing point source categories. These nine categories account for over 99% of the 1976 Canadian HC and NO_x emissions.

TABLE 2. MAJOR CANADIAN POINT SOURCES CATEGORIES

Category	Emissions % NO_x	 % HC
Petroleum refining	12	95
Natural gas production	26	2
Electric power generation	52	2
Nitric acid production	2	≃0
Tar sands operations	2	<1
Sulphate pulping	4	≃0
Manufacture of carbon and graphite electrodes	1	<1

Seasonal data were obtained for NO_x and SO_x only. Therefore, any further temporal resolution is currently provided by applying the temporal factors developed for the U.S. inventory to appropriate source categories.

Speciation of Canadian HC data can be accomplished by using the U.S. methodologies where appropriate correlations can be made between U.S., SCC, and Canadian SIC. Additional profiles may be introduced to define more realistically the chemical species associated with the Canadian processes.

Most point source emissions data supplied by the provincial agencies and regional offices are obtained from individual companies through stack tests, engineering analyses, or mass balance methods. Most other emission estimates are developed by using standard U.S. emission factors (EPA, 1977) and either plant-specific or general fuel use production and consumption data.

AREA SOURCE DATA

U.S. Inventory

Area sources comprise stationary and mobile sources that typically emit less than 100 tons/yr and are too small and/or too numerous to record individually. Many states submitted their current area source inventory either in hard copy or in computer format compatible with EIS/AS. Standard methodologies (EPA, 1982) were recommended to ensure consistency from state to state. Annual county emissions of VOCs and NO_x were reported for the 54 area source categories shown in Table 3. Raw data from 1970 to 1980, which are used in the annual county emission calculations, are extremely disparate: gasoline and fuel oil sales; industry employment levels; county population; asphalt paving; diluent content of asphalt; pesticide use or acres harvested; highway vehicle classes and vehicle miles traveled; fuel consumption; registration data; vessel movement; and agricultural, highway, and business statistics. Because of their significant contribution (62% of NO_x emissions and 48.3% of VOC emissions), highway mobile sources divided into the following categories: light-duty vehicles (LDV); light-duty gasoline-powered trucks, 0 to 6,000 lbs GVW (LDT1); light-duty gasoline-powered trucks, 6,000 to 8,500 lbs GVW (LDT2); heavy-duty gasoline-powered trucks (HDG); heavy-duty diesel-powered vehicles (HDD); and motorcycles. For some of the states, area source inventories were either unavailable or sufficiently out of date or incomplete, so that they had to be replaced by contractor-generated data by the procedures recommended above.

Annual county emissions for the 54 source categories were apportioned to the previously defined grid system to satisify the spatial requirements of regional-scale modeling. Spatial apportionment factors were developed to allocate a portion of a particular county's area emission to a specific grid cell according to the known distribution of some surrogate indicator. Surrogate indicators used in NECRMP are: housing, population, urban land, agricultural land, composite forest, land area, airport location, and park location. The particular surrogate indicator assigned for allocation of emissions in each area source category is listed in Table 3. Distribution of the surrogate indicators over the NECRMP grid was obtained from various U.S. Bureau of the Census statistics, land use classification data derived from Landsat imagery, the U.S. Geological Survey maps, and Waterborne Commerce of the United States - 1978. A homogeneous distribution of land use within a grid was assumed.

Temporal distribution of annual area source emissions is achieved by applying seasonal, daily, and hourly factors for each source category. The standard algorithm described in the U.S. point source section is applied. Table 3 summarizes the temporal patterns used for the NECRMP emissions. Details and comprehensive source references for developing spatial and temporal allocation factors are listed at the end of this paper (EPA, 1983, 1979, 1978).

TABLE 3. NECRMP TEMPORAL ALLOCATION PATTERNS AND SPATIAL ALLOCATION SURROGATES FOR AREA SOURCE CATEGORIES

Category Description	Surrogate Indicator	Seasonal	Daily	Hourly
Stage I Gasoline Evap.	2	uniform	Mon-Sat	0600-2000
Stage II Gasoline Evap.	2	uniform	7 dys/wk	varies each hour (see Ref. 2)
Storage Tank Breathing	2	uniform	7 dys/wk	all hours
Gasoline Loading/Transit	2	uniform	Mon-Sat	0600-2000
Sm Ind/Comm Degreasing	3	uniform	Mon-Sat	80% from 0700-1800, 20% from 1900-2400
Dry Cleaning	2	uniform	Mon-Sat	0700-1800
Arch Surface Coating	3	uniform	7 dys/wk	0600-2000
Auto Body Refinishing	3	uniform	Mon-Sat	0800-1800
Sm Ind Surface Coating	3	uniform	Mon-Sat	0700-1900
Graphic Arts	2	uniform	Mon-Sat	75% from 0800-1800; 25% from 1900-0700
Comm/Cons Solvent Use	2	uniform	7 dys/wk	75% from 0600-1700; 25% from 1800-0500
Cutback Asphalt	3	zero in winter, uniform over other seasons	7 dys/wk	all hours
Pesticides	4	state specific	7 dys/wk	0700-1900
On-Highway LDV	2	uniform	7 dys/wk	varies each hour (see Ref. 1)
On-Highway LDT1	2	uniform	7 dys/wk	
On-Highway LDT2	2	uniform	7 dys/wk	
On-Highway HDG	2	uniform	7 dys/wk	
On-Highway HDD	2	uniform	7 dys/wk	
On-Highway MC	2	uniform	7 dys/wk	
Residential Anthracite	1	Based on monthly average degree-day data at a representative meteorological site in each state. (see Ref. 7)	7 dys/wk	Based on monthly averages of 3 hour met. records; season specific for each state. (see Ref. 7)
Residential Bituminous	1		7 dys/wk	
Residential Residual Oil	1		7 dys/wk	
Residential Distillate	1		7 dys/wk	
Residential Natural Gas	1		7 dys/wk	
Residential LPG	1		7 dys/wk	
Residential Wood	1		7 dys/wk	

TABLE 3. (continued)

Category Description	Surrogate Indicator	Seasonal	Daily	Hourly
Comm/Inst Anthracite	2	35% winter, 25% spring, 15% summer, 25% fall	Mon-Sat	80% from 0800-2300; 20% from 2400-0700
Comm/Inst Bituminous	2	• • • •	•	• •
Comm/Inst Residual Oil	2	• • • •	•	• •
Comm/Inst Distillate Oil	2	• • • •	•	• •
Comm/Inst Natural Gas	2	• • • •	•	• •
Comm/Inst LPG	2	• • • •	•	• •
Comm/Inst Wood/other	2	• • • •	•	• •
Industrial Anthracite	3	uniform		• •
Industrial Bituminous	3	uniform	•	50% from 2000-0700; 50% from 0800-1600
Industrial Residual Oil	3	uniform	•	• •
Industrial Distillate Oil	3	uniform	•	• •
Industrial Natural Gas	3	uniform	•	• •
Industrial LPG	3	uniform	•	• •
Industrial Wood/other	3	uniform	•	• •
Military Aircraft	7	uniform	7 dys/wk	45% from 2100-0800; 55% from 0900-2000
Civil Aircraft	7	uniform	7 dys/wk	0700-2100
Commercial Aircraft	7	25% winter, 23% spring, 26% summer, 26% fall	7 dys/wk	90% from 0700-2400; 10% from 0100-0600
Railroad Locomotives	6	uniform	Mon-Sat; Sun (1/3 dy)	70% from 0700-1800; 30% from 1900-0600
Vessels-Gasoline	8	50% summer, 50% uniform over spring & fall	7 dys/wk	0700-1800
Vessels-Distillate Oil	8	uniform	7 dys/wk	75% from 0700-1900; 25% from 2000-0600
Vessels-Residual Oil	8	uniform	7 dys/wk	75% from 0700-1900; 25% from 2000-0600
Off-Highway Vehicle-Gas	5	state specific (see Ref. 7)	7 dys/wk	0 from 2000-0600; 4% @0700, 4.5% @0800, 6.2% @0900, 7% @1900, 78.3% @1000-1800
Off-Highway Vehicle-Diesel	5	state specific (see Ref. 7)	7 dys/wk	0 from 0100-0600; 1.4% @0700, 93.6% from 0800-1900, 5% from 2000-2400
On-site Incineration	2	uniform	5 1/2 dys/wk	0700-1700
Open Burning	6	uniform	7 dys/wk	0600-2000
Structural Fires	1	uniform	7 dys/wk	all hours
Field/Slash Burning	4	10% winter, 70% spring, 0% summer, 20% fall	7 dys/wk	0500-1900
Forest Fires	5	5% winter, 5% spring, 50% summer, 40% fall	7 dys/wk	all hours

- distribution assumed region-wide unless specified
- distribution uniform over specified periods unless stated; zero otherwise
- local time; hour specified designates the ending time of the hour period (i.e., 0800 implies the period between 0700 and 0800).

Volatile organic compound emissions for area sources are estimated by using the same methodology as that discussed for point sources, by assigning a pseudo SCC to each area source category.

The spatial, temporal, and speciation factors are appropriately applied to the annual source emissions through the use of the Regional Model Data Handling System (RMDHS) (EPA, 1981), a COBOL software package specifically designed to interface with the ROM.

Canadian Inventory

Canadian area sources consist of minor point sources, mobile sources, and other sources too small or too numerous to track individually. Emission totals have been calculated for 54 SICs. However, these do not correspond directly to the 54 U.S. area source categories. The following major categories account for over 85% of both NO_x and HC emissions in Canada: diesel engines, diesel and gas marketing, forest fires, fuel combustion, and general-purpose motor vehicles. The Canadian National Emissions Inventory available to the U.S. currently does not record area source emissions by source category. Only total emissions for each pollutant are reported on the polar stereographic grid system used by the Canadian Meteorological Center (CMC). The side length of a grid cell is 127 km, and each grid is identified by the x-y coordinates of the south-west corner. Latitude and longitude information is also available. Emissions are reported on the CMC grid system for all of Canada; however, only those portions falling within the NECRMP region are included in the modeling inventory. Written communication from A. Sheffield, Air Pollution Control Directorate, Environment

Canada, Ottawa, Ontario, provided the percentage of contribution for each area source category. Currently, this source distribution and a surrogate indicator of population by census district are the only data available for source category and spatial apportionment. More detailed source and spatial distribution data such as population, housing counts, and fuel type by enumeration district, employment statistics, and industry location data will be available from Environment Canada in the future. When appropriate, the U.S. temporal apportionment and VOC speciation methodologies will be applied to obtain the data resolution required for Eulerian regional-scale models.

DATA QUALITY

An emissions inventory is a compilation of estimates. As such, it does not possess the capability of comparison to true and firm standards. Nevertheless, if standard, proven procedures are followed and the resulting data are subjected to logical quality assurance and validation checks, one can develop a reasonable assurance that an inventory data base is reliable for the purposes intended.

The NECRMP inventory is being composed of input from several states and other jurisdictions that has existed in several, often incompatible formats. Thus, an early stage of the data compilation and quality assurance effort was to reduce the data to a common format. The NEDS/EIS format was chosen. (NEDS and EIS are compatible, and output from one may generally be used to update the other.) For most states in the NECRMP area, the reduction to a common format for point sources was done either largely or entirely by computer. Therefore, as the data were processed into a standard format, they were edited and became amenable to further computerized logic checks and validation procedures.

The following list is fairly typical of the checks that were done during the edit step.

- Determine whether valid numeric characters are in numeric fields.
- Check UTM coordinates against a range of possible valid values for the state.
- Flag excessive values when compared with expected norms.
- Cross-check for specific missing data.
- Cross-check information on stack height, diameter, plume rise, temperature, velocity, boiler firing rates, etc.

Several programs were developed that supplement the EIS edit and summary reporting capabilities, and provide "fixes" to the various inventory files. These programs produce files and format printouts that organize the data into a form whereby easily managed manual checking and "common or generic" error repairs can be performed.

The NEDS-to-EIS-conversion editor program and supplemental programs that were developed provided lists of all severe and conditional warning errors. Errors considered important for NECRMP modeling were documented for submittal to the states. Similar listings were generated from the master file creation program checks, which identified such problems as duplicate transactions and incomplete records. Perhaps one of the most valuable steps was the manual review of the major point sources. All sources with VOC or NO_x emissions estimated at ≥500 tons/yr were so reviewed. Inconsistencies, nonsensical entries, engineering unreasonableness and inaccuracies, etc., were indentified and flagged for further investigation and resolution. External data bases (e.g., lists of all sources in an industry category as available from a trade association listing) were used to cross-check and identify missing source possibilities.

Upon completion of this review and consolidation of all questions and identified errors, the lists and supporting information were forwarded to the states for resolution. The state agencies reviewed the problem lists and searched their records for answers. In most instances, the contractor and the state personnel met to assure that the problems and responses were properly communciated. Many telephone calls were made to follow up individual problems. In some instances, the contractor also spot-checked the agency's file data to gain an added measure of understanding and confidence in the completeness and accuracy of the data.

After revising the data base to add, delete, or correct information found in error, the data were then reprocessed through the system to ensure that the updates were done properly and that the same or new errors did not exist. Resulting data were made available to the states for their use in the normal updating of NEDS, SIPs, etc.

Similar procedures were followed for the area source data base. However, there was one major distinction in that the states in many cases did not maintain a current area source data base. In these cases, the contractor compiled the data according to standardized procedures (EPA. 1980, 1982).

Thus, the NECRMP emissions inventory is believed to be as complete as is practical under the resources available and within the realm of current responsibilities. Undoubtedly, errors remain and some sources may be omitted. Since the data base can be continually updated, it is likely that some additional corrections may be warranted in the future. For example, through the modeling process itself, peculiar results will often surface that will culminate in the identification of a missing source or erroneous information on an included source.

As alluded to earlier, the estimated data are for 1979/1980 (or at least adjusted to that time period). Inventories are of necessity retrospective (unless projected in some manner). Thus, the data compiled in NECRMP are as appropriate for the time period of estimate as can now be determined.

In summary, an emissions inventory can never be proven precise and accurate. The NECRMP inventory, however, has undergone rigorous checks and validation efforts and is believed to be capable of driving the ROM. Any errors or inconsistencies identified in the future can be subsequently corrected.

The Canadian inventory is as complete as possible in terms of major source contributions. However, the lack of detailed data for determining temporal distribution of point and area annual emissions affects the accuracy of the hourly resolved emissions estimates. The use of U.S. factors, when appropriate, assuages that effect. The aggregate nature of the reported area source emissions definitely weakens any speciation or spatial allocation methodologies. Future updates must concentrate on providing more resolution in these areas, particularlv for major contributing source categories.

The currentness of Canadian emissions data varies by pollutant. Data for particulates, CO, and HC from petroleum refineries are reported for 1976. Data for NO_x, SO_x, and the remaining HC emissions have been updated to 1978. A few additional updates for 1980 have been incorporated into the National Canadian Emissions Inventory. Further updates toward a 1980 base year will be incorporated into the NECRMP inventory when received from Environment Canada.

The Environmental Protection Service (EPS) of Canada maintains the quality of the emissions inventories. However, since there are no direct reporting requirements, the province-supplied data are typically accepted unless a large discrepancy is noted. EPS compiles, reviews, and processes via computer the emissions data that ultimately become part of the Canadian National Emissions Inventory.

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DISCUSSION

S. Reynolds: What, if anything, has EPA done to quantify the uncertainties in the emissions inventory?

J. Novak: There is a report out. I am not totally familiar with everything that was done in terms of quantification. All I can say is that the procedures that were undertaken were done to actually verify the comprehensiveness. Joe Southerland was responsible for that inventory development. Maybe he could better answer that question.

J. Southerland: The job of attaching some statistical uncertainty to an inventory is a very difficult, if not impossible, task. A number of studies and papers have looked at this kind of thing. Essentially, it comes down to the fact that an inventory is kind of an iterative process. It is dynamic in that you are always finding additional uncertainties or errors. For some of the things, like an omitted source, how do you quantify the degree of uncertainty when you do not realize that the source is there?

So, for this particular inventory, we have a report in draft form. It is essentially an evaluation document that will look at the improvements we have made in the inventory and the uncertainties of the unknowns, some of the things that we except or suspect to be additional things that could be improved in the inventory. It is a kind of evaluation document that will enable additional steps to be taken if so desired to continue this iterative process.

L. Lindau: I think we will come back to these problems in the general discussion. I think this is a very important question.

E. Runca: Can you define what you mean when you say that these data can be used to develop emission scenarios? Are these projections in the future?

J. Novak: In my understanding of what types of emission scenarios would want to be developed, possibly certain source categories would be earmarked for reduction. For instance, you have 14-area 54-source categories. Highway sources would be a major contributor we might like to reduce. This system has the flexibility such that factors could be put in if you wanted a 50% reduction or 20% reduction: The entire system could be executed and run to produce an emission scenario that then included reduced highway sources or whatever other combinations of reductions for individual sources that you wanted. You could come up with various strategies for how you are going to control certain source types, develop the strategies, put them into the system, and come out with emission scenarios that would reflect those control strategies.

E. Runca: Are economic and technological factors taken into account?

J. Novak: No. They would have to be taken into account in terms of inputting the appropriate reduction. This is not an economic analysis; it is strictly a software package to produce the required emissions to the right levels for the different sources.

EMISSION INVENTORY DATA BASES FOR THE UNITED STATES*

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INTRODUCTION

In the United States, most emissions inventory activity has been performed by state and local air pollution control and planning agencies. These agencies develop emissions inventories to support the State Implementation Plans (SIPs) required by the U.S. Clean Air Act. Point source emissions inventory data are required by U.S. Environmental Protection Agency (EPA) regulations, to be submitted by the states to EPA in the format of the National Emissions Data System (NEDS). The NEDS data base is thus the most readily available emissions inventory information in the United States.

NEDS has been in existence since 1972. Currently, it contains data for about 55,000 establishments defined as point sources. NEDS also reports estimated emissions for all other sources not represented in the point source file as area source emissions. The data in the point and area source files are described briefly below.

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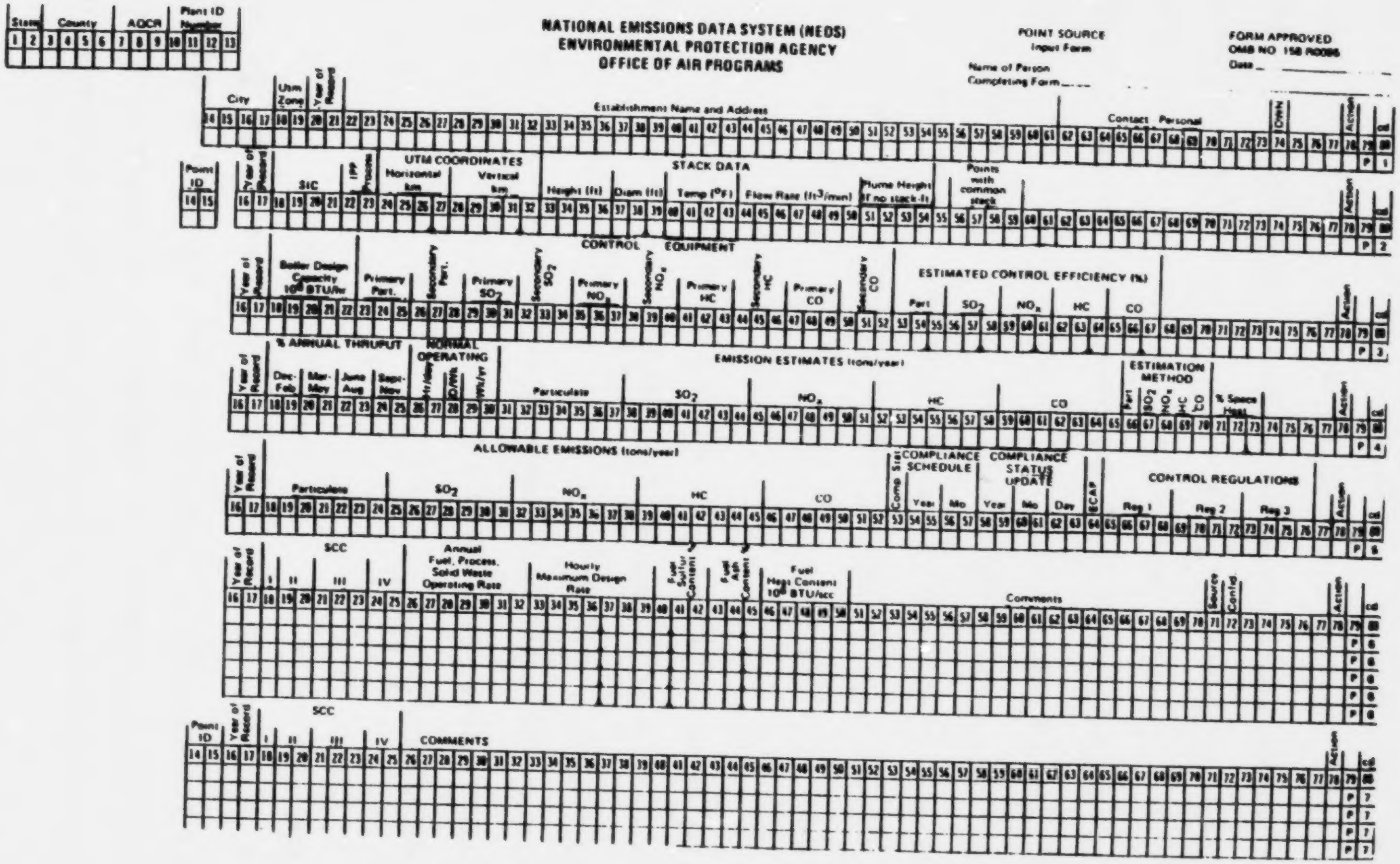
*This paper has been reviewed by the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

NEDS POINT SOURCE DATA

NEDS point source data are all submitted to EPA by state and local air pollution agencies. EPA regulations require that data be submitted on an annual basis for all establishments with annual actual emissions of more than 100 tons (90.7 metric tons) of particulate matter, SO_2, NO_2, or VOCs or more than 250 tons (227 metric tons) of CO. Most states report more data than are actually required. Only about 13,000 of the 55,000 establishments currently in NEDS actually emit over 100 tons per year. All 50 states plus the District of Columbia, Puerto Rico, the Virgin Islands, and Guam submit point source data. Data are collected by each state using its own procedures. Most rely upon a combination of source permit systems, emissions inventory questionnaires, and on-site facility inspections to obtain data.

Point source data collected by a state are normally maintained in an automated data system by the state agency. About 20 agencies use the Emissions Inventory System (EIS) designed by EPA specifically for state use. Approximately 15 other agencies use other automated systems. The remaining agencies, usually those with a relatively small number of sources, maintain their records manually.

Data are submitted to EPA in the standard NEDS format, which is illustrated in Figure 1. The basic data elements are the establishment name and address, the Standard Industrial Classification (SIC) code, the Universal Transverse Mercator (UTM) grid coordinates, stack parameters, control equipment data, source operating schedules, source operating rates and fuel characteristics, and



NATIONAL EMISSIONS DATA SYSTEM (NEDS)
ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF AIR PROGRAMS

POINT SOURCE
Input Form

Name of Person
Completing Form ____

FORM APPROVED
OMB NO 158 R0086
Date ____

State | County | AQCR | Plant ID Number

City | Utm Zone | Year of Record | Establishment Name and Address | Contact Personal | OWN | Action | cd | P 1

Point ID | Year of Record | SIC | IPP | Process | UTM COORDINATES Horizontal km | Vertical km | STACK DATA Height (ft) | Diam (ft) | Temp (°F) | Flow Rate (ft³/min) | Plume Height If no stack-ft | Points with common stack | Action | cd | P 2

CONTROL EQUIPMENT: Year of Record | Boiler Design Capacity 10⁶ BTU/hr | Primary Part. | Secondary Part. | Primary SO₂ | Secondary SO₂ | Primary NOₓ | Secondary NOₓ | Primary HC | Secondary HC | Primary CO | Secondary CO | ESTIMATED CONTROL EFFICIENCY (%) Part | SO₂ | NOₓ | HC | CO | Action | cd | P 3

Year of Record | % ANNUAL THRUPUT Dec-Feb | Mar-May | June-Aug | Sept-Nov | NORMAL OPERATING Hr/day | D/Wk | Wk/yr | EMISSION ESTIMATES (tons/year) Particulate | SO₂ | NOₓ | HC | CO | ESTIMATION METHOD Part | SO₂ | NOₓ | HC | CO | % Space Heat | Action | cd | P 4

Year of Record | ALLOWABLE EMISSIONS (tons/year) Particulate | SO₂ | NOₓ | HC | CO | Comp Stat | COMPLIANCE SCHEDULE Year | Mo | COMPLIANCE STATUS UPDATE Year | Mo | Day | ECAP | CONTROL REGULATIONS Reg 1 | Reg 2 | Reg 3 | Action | cd | P 5

Year of Record | SCC I | II | III | IV | Annual Fuel, Process, Solid Waste Operating Rate | Hourly Maximum Design Rate | Fuel Sulfur Content | Fuel Ash Content | Fuel Heat Content 10⁶ BTU/scc | Comments | Source | Confid | Action | cd | P 6

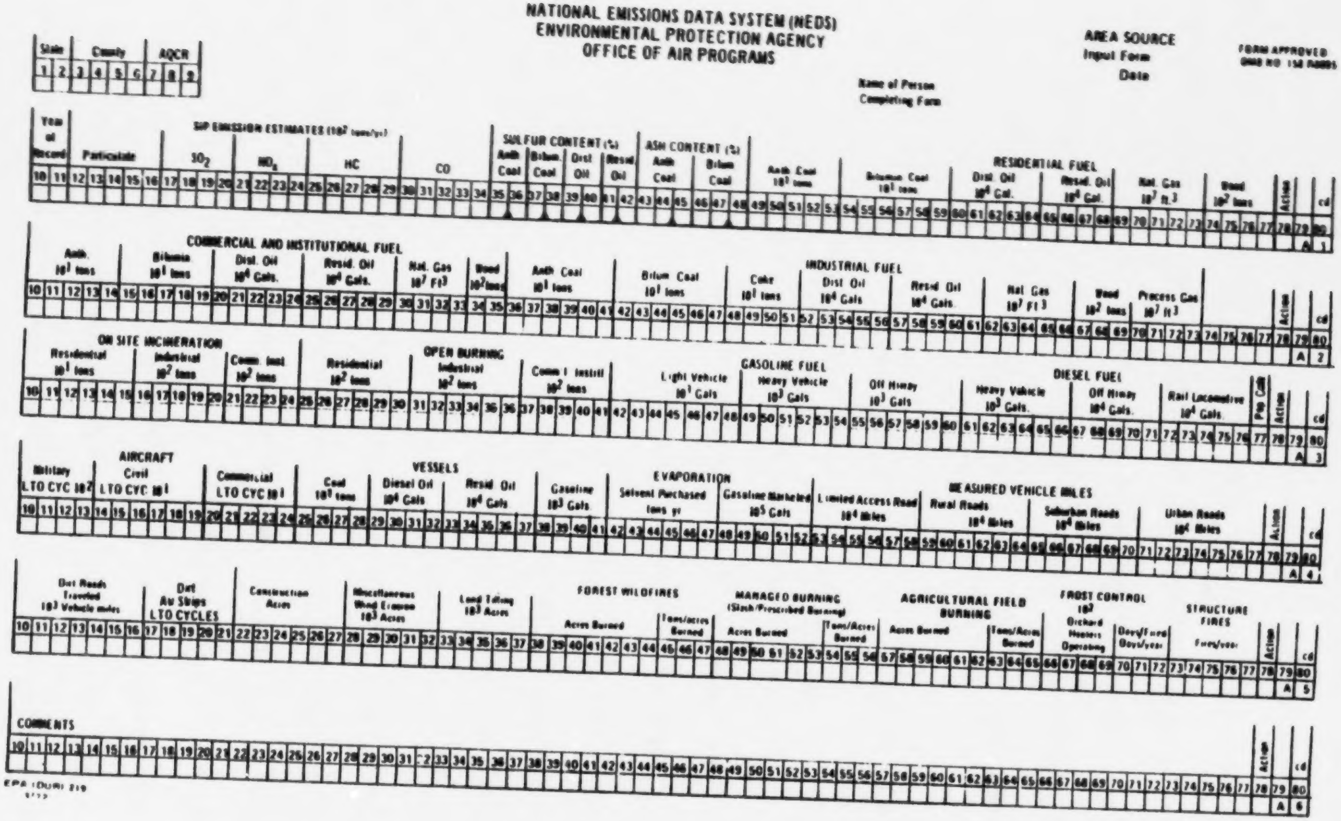
Point ID | Year of Record | SCC I | II | III | IV | COMMENTS | Action | cd | P 7

Figure 1. NEDS point source form.

estimated emissions. In NEDS, all emissions are reported as annual actual emissions estimates. These estimates, which may be based on stack results, material balance calculations, or special state-defined emission factors, may be hand calculated and entered by the state agency. Alternatively, the state may specify that emissions be calculated by EPA by using a standard file of emission factors along with reported source operating rates and control efficiency data. These standard emission factors are available in Compilation of Air Pollution Emission Factors (EPA, 1978).

NEDS AREA SOURCE DATA

In NEDS, the county or county equivalent in each state is represented as an area source. Thus, about 3,200 area source records are maintained in NEDS. Area source data are updated annually by EPA. States are not required to submit area source data, and very little data are voluntarily submitted. EPA develops area source emissions estimates based on the standard NEDS area source categories shown on the NEDS area source form (Figure 2). For each of these categories, county-level estimates of source activity are derived. These are normally based on data published by other Federal agencies, such as the Department of Energy, Department of Transportation, etc. Often, needed data are available only at the state level. EPA has developed a set of procedures to allocate state-level data to individual counties, based on the use of population, employment, or other data that are available at the county level (EPA, 1978). All county-level estimates of area source activity are converted to emissions by using standard emission factors. For most source categories, these data are taken from the EPA reference document cited above (EPA, 1978).



NATIONAL EMISSIONS DATA SYSTEM (NEDS)
ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF AIR PROGRAMS

AREA SOURCE
Input Form
Date

FORM APPROVED
OMB NO. 158 R0005

Name of Person
Completing Form

State	County	AQCR
1 2	3 4 5 6	7 8 9

Year of Record		10–11
SIP EMISSION ESTIMATES (10^2 tons/yr)	Particulate	12–16
	SO_2	17–20
	NO_x	21–24
	HC	25–29
	CO	30–34
SULFUR CONTENT (%)	Anth Coal	35–36
	Bitum. Coal	37–38
	Dist Oil	39–40
	Resid Oil	41–42
ASH CONTENT (%)	Anth Coal	43–45
	Bitum Coal	46–48
RESIDENTIAL FUEL	Anth Coal 10^3 tons	49–53
	Bitumin Coal 10^3 tons	54–59
	Dist. Oil 10^4 Gal.	60–64
	Resid. Oil 10^4 Gal.	65–68
	Nat. Gas 10^7 ft^3	69–73
	Wood 10^2 tons	74–77
	Action	78
		79 A
	cd	80 1

COMMERCIAL AND INSTITUTIONAL FUEL	Anth. 10^1 tons	10–14
	Bitumin 10^1 tons	15–19
	Dist. Oil 10^4 Gals.	20–24
	Resid. Oil 10^4 Gals.	25–29
	Nat. Gas 10^7 Ft^3	30–33
	Wood 10^2 tons	34–35
INDUSTRIAL FUEL	Anth Coal 10^1 tons	36–41
	Bitum Coal 10^1 tons	42–47
	Coke 10^1 tons	48–51
	Dist Oil 10^4 Gals	52–56
	Resid Oil 10^4 Gals.	57–61
	Nat Gas 10^7 Ft^3	62–66
	Wood 10^2 tons	67–69
	Process Gas 10^7 ft^3	70–73
		74–77
	Action	78
		79 A
	cd	80 2

ON SITE INCINERATION	Residential 10^1 tons	10–15
	Industrial 10^2 tons	16–20
	Comm. Inst 10^2 tons	21–24
OPEN BURNING	Residential 10^2 tons	25–30
	Industrial 10^2 tons	31–36
	Comm'l Instit'l 10^2 tons	37–41
GASOLINE FUEL	Light Vehicle 10^1 Gals	42–48
	Heavy Vehicle 10^3 Gals	49–54
	Off Hiway 10^3 Gals	55–60
DIESEL FUEL	Heavy Vehicle 10^3 Gals.	61–66
	Off Hiway 10^4 Gals.	67–71
	Rail Locomotive 10^4 Gals.	72–76
	Pop Cat	77
	Action	78
		79 A
	cd	80 3

AIRCRAFT	Military LTO CYC 10^2	10–13
	Civil LTO CYC 10^1	14–19
	Commercial LTO CYC 10^1	20–24
VESSELS	Coal 10^1 tons	25–28
	Diesel Oil 10^4 Gals	29–32
	Resid Oil 10^4 Gals	33–37
	Gasoline 10^3 Gals	38–41
EVAPORATION	Solvent Purchased tons yr	42–47
	Gasoline Marketed 10^5 Gals	48–52
MEASURED VEHICLE MILES	Limited Access Road 10^4 Miles	53–58
	Rural Roads 10^4 Miles	59–64
	Suburban Roads 10^4 Miles	65–70
	Urban Roads 10^4 Miles	71–77
	Action	78
		79 A
	cd	80 4

	Dirt Roads Traveled 10^3 Vehicle miles	10–16
	Dirt Air Strips LTO CYCLES	17–21
	Construction Acres	22–27
	Miscellaneous Wind Erosion 10^3 Acres	28–32
	Land Tilling 10^3 Acres	33–37
FOREST WILDFIRES	Acres Burned	38–44
	Tons/acres Burned	45–47
MANAGED BURNING (Slash/Prescribed Burning)	Acres Burned	48–53
	Tons/Acres Burned	54–56
AGRICULTURAL FIELD BURNING	Acres Burned	57–62
	Tons/Acres Burned	63–65
FROST CONTROL	10^2 Orchard Heaters Operating	66–69
	Days/Fired Days/year	70–72
STRUCTURE FIRES	Fires/year	73–77
	Action	78
		79 A
	cd	80 5

COMMENTS		10–77
	Action	78
		79 A
	cd	80 6

EPA (DUR) 219

Figure 2. NEDS area source form.

Highway vehicle emission factors for each county are calculated by using a simplified version of the MOBIL2 model (EPA, 1981), which takes into account vehicle age distribution, applicable emission standards, average vehicle speeds, average ambient temperature, and other factors. As with point source data, area source emissions in NEDS are reported only as annual actual emissions.

AVAILABILITY OF NEDS DATA

The NEDS point and area source files reside in EPA's Univac computer at Research Triangle Park, North Carolina. Data are available in a variety of standard computer printout formats or on magnetic tape files (EPA, 1980). In addition, qualified EPA Univac users may access the files directly. However, because of confidentiality claims made by states, free access to the NEDS point source file is not granted to all potential users. In addition, data items identified as confidential are excluded from computer printouts and data files made available to non-EPA users. Data items sometimes identified as confidential are source operating rates, capacities, and emission estimation method codes that, if revealed, would possibly allow calculation of confidential source operating rates. No emissions estimates can be claimed as confidential, however, under the provisions of the Clean Air Act.

OTHER DATA BASES

Recent activities in photochemical oxidant and acid deposition research have promoted the development of a number of other emissions data bases. Many of these have been developed by using the existing NEDS data as a starting point to improve the data by additional data collection or additional software to process the data. These data bases have been reviewed in another paper (Bosch, 1982). A few of these other data bases are briefly identified below.

Northeast Corridor Regional Modeling Project (NECRMP) Inventory

These data were developed for 15 Eastern United States to provide input to photochemical oxidant models and will eventually be included in NEDS. This project will be discussed further in a separate presentation at this conference.

Sulfate Regional Experiment (SURE)

These data were developed by the Electric Power Research Institute (EPRI) for use in sulfate episode modeling, long-term transport of sulfates, and the Utility Simulation Model. The data base covers the Eastern United States, emphasizing emissions of SO_x and related species for the period 1977 to 1978. EPRI is currently sponsoring a major new effort to develop emissions data for a 1982 base year (Heisler, 1982). These data are to include emissions estimates for SO_2, sulfates, NO, NO_2, total particulates, and VOCs by reactivity class for the United States, excluding Alaska and Hawaii. Seasonal and daily average emissions estimates are also to be included.

Hazardous and Trace Emissions System (HATREMS)

This is a companion system to NEDS that is maintained by EPA for calculating emissions estimates of other pollutants not included in NEDS.

Basically, HATREMS provides a separate emission factor file that may be used to calculate emissions by using NEDS source parameters. Presently, HATREMS is routinely used only for reporting lead emissions.

Multistate Power Production Pollution Study (MAP-3S)

These data were developed by Brookhaven National Laboratory to provide data for air quality forecasting and econometric models. The data base started with 1976 base-year NEDS data and has been upated to include data for major Canadian sources, as provided by Environment Canada and improvements for the SURE data base. The data now represent a 1978 base year for annual emissions of particulates, SO_2, NO_2, VOCs, and CO.

Historical Trends

National trends in the emission of particulates, SO_2, NO_2, VOCs, and CO have been reported to EPA (1982a,b). These data are intended primarily to identify, on a national basis, the long-term trends in emissions as well as recent changes caused as the result of air pollution control efforts. These reports show trends in emissions by source category for general management and public information purposes. More detailed historical emissions estimates have been developed for SO_2 and NO_2 (Gschwandtner et al., 1981). State-level estimates of these emissions for major source categories have been developed for the period 1950 to 1978 for states in the Eastern United States. Work is now in progress to expand the data base to include all states and all years, in 5-yr intervals, for the period 1900 to 1980.

CURRENT DEVELOPMENTS

To meet the needs of the Federal National Acid Precipitation Assessment Program (NAPAP), EPA is developing a new emissions inventory data base. The data are being installed in an Emissions Inventory System file on EPA's Univac computer at Research Triangle Park, North Carolina. The data base will represent the base year 1980 and will cover all states except Alaska and Hawaii. The initial data file has been created from current NEDS data. These data are being improved by using input from the users and quality assurance activities conducted by the data base manager. Data for major Canadian sources are also being included in the file.

The objective of creating the data base is to provide a single, common set of 1980 base-year data to be used as the starting point for acid deposition related analyses requiring emissions data. Atmospheric modeling activities involving the development of both Lagrangian and Eulerian transport models will be supported. The data base may be expanded to include additional pollutants such as sulfates, ammonia, and chlorides as required. Thorough documentation of the NAPAP inventory is planned. Updates to the data base will be subject to peer review and must meet the needs of the models, procedures for temporal and spatial allocation of emissions will be developed. A procedure for allocating VOC species into reactivity classes, drawing upon the information available from the NECRMP project, is also planned. These efforts are expected to take place over the next few years. Plans also call for the eventual development of a 1984 base-year inventory by FY1988. Further information on the NAPAP data may be obtained by contacting the data base administrator, David Mobley, at 919-541-2578 (FTS 629-2578) or the author of this paper.

NEDS is expected to continue normal operation for at least 2 yr. EPA is developing a new data base management system, the Aerometric Information Retrieval System (AIRS), that is intended to replace NEDS, HATREMS, EIS, and other systems for storing air quality data. AIRS is scheduled to be completed in 1985 at projected budget levels. When completed, AIRS will make available to state and local agencies, as well as to EPA users, a more advanced state-of-the-art software system for processing emissions and air quality data.

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DISCUSSION

P. Misra: Could you explain the differences between the different emissions inventories, such as SURE, NECRMP, and RTS?

C. Mann: I am not sure I can explain the differences in a brief time period. Each of the data files was developed to serve a slightly different purpose. They started at different points in time, using different NEDS files, and changes were made in each inventory based on different data that were collected, different assumptions that were made, and different emission factors that were used. Technically, there are a number of reasons why these things are different.

One of the things that we are trying to avoid in creating the NAPAP file is the proliferation of multiple sets of data bases such as those people have been using in the past. Ideally, we want one single set of data for everybody to start with as the basic emissions inventory data that goes into their analysis.

EMISSIONS INVENTORIES AND THE NATIONAL EMISSIONS INVENTORY SYSTEM*

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INTRODUCTION

The National Emissions Inventory System (NEIS) is a data storage and retrieval system maintained by the Inventory Management Division, Program Management Branch of Environment Canada. Such an inventory of sources and emissions is a prerequisite to any national program on air pollution and abatement. It provides the information that enables problem areas to be identified and priorities to be set. The information identifies and locates all sources, types, and quantities of emissions. Although an emissions inventory may be regarded as simply an information storage, processing, and retrieval system, it is nonetheless the most important planning tool in a comprehensive program of air pollution control. The data that a complete, up-to-date inventory provides may be used, for example, in the design of an air sampling network to predict ambient air quality in a region or to evaluate or modify a control program.

Assessing national air pollution requires accurate data on the quantity and characteristics of emissions from all sources that contribute to the problem. The number and the diverse types of sources make field measurements of emissions on a source-by-source basis impractical. Other means for collecting emissions data for each source include: questionnaires, permits, telephone or written communications, and individual company records. When specific information is not available through any of these means, data from published reports dealing with production and/or consumption statistics and emission factors are used.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

Data of this nature are obtained from various sources, such as company-specific information from questionnaires. However, most information is collected from various Federal departments (e.g., Statistics Canada) or from provincial regulatory agencies, as in the case with permit data. In certain situations, data are collected by private consultants who may obtain their information from any of the above-mentioned sources.

Data in NEIS are updated every 2 yr and are summarized in the publication, A Nationwide Inventory of Emissions of Air Contaminants. These updates are not required under law, but they are performed for the purpose of comparing emissions data on a year-to-year basis. In order to preserve confidentiality, data are summarized in the publication; emissions for individual plants are not given. The same restriction does not necessarily apply to requests for information from the NEIS. The release of data depends upon the source of the request. That is, in the case of public or private consultants, data are "rolled up" in order to maintain the rules of confidentiality, but plant names may be given to, for example, Environment Canada regional personnel.

The inventory covers the entire country (i.e., 10 provinces and 2 territories) and five major source categories: industrial processes, stationary source fuel combustion, transportation, solid waste incineration, and miscellaneous. Emissions from approximately 80 sectors are calculated and can be resolved spatially in various ways, including a 127 km x 127 km grid (derived from latitude/longitude coordinates).

POINT SOURCE DATA

A point source can be defined as a plant consisting of a number of stacks and any number of processes. The exact location of the plant and site-specific data are available for these sources.

Raw Data

Within the hierarchy of NETS, various levels contain detailed information on the plant: specific location, stack information, process description, and emissions.

Available stack information consists of the following: height, diameter, temperature of the flue gas at the point of exit, flow rate of flue gas, exit velocity of the flue gas, and dust concentration.

The industries contributing the most to the pollution burden vary according to the contaminant under consideration. Based on the 1978 inventory, the major contributing industrial sources for each contaminant were as follows:

- Particulate matter--iron ore mining and beneficiation, mining, rock quarrying, and power plants;
- SO_2--primary copper and nickel production power plants and natural gas processing;
- NO_x--power plants, sulphate pulping, and tar sands operations;
- HCs--petrochemical industry, petroleum refining, and crude oil production; and
- CO--petroleum refining, iron and steel production, and carbon black.

The types of raw data that are collected vary. For example, plant production or capacity are usually available from various sources. Other types of data that are collected include: fuel consumption, electrical generation in the case of power plants, sulphur/ash content of fuels, operating schedules, pollution control equipment and their efficiencies, stack information, emission factors, and actual emissions. The temporal resolution of these data is usually annual but at times is seasonal (quarterly). Point source data, if received through questionnaires, may contain latitude/longitude coordinates, thus enabling in-house staff to calculate grid coordinates by using a software package. Other forms of its location (e.g., city or census division) are easily determined.

Data are available on the NEIS for 1974, 1976, and 1978; major point soures have been updated to 1980. Additional information for 1970 and 1972 is available in hard copy.

Emissions Data

The contaminants for which data are stored in the NEIS were discussed in the previous section, with the exception of primary sulphates. In addition, emissions data for particulate matter are distributed by particle size, and HCs are available on a compound class basis. Emissions data for particulate matter are divided into three distributions: <2.5 μm, 2.5 to 15 μm, and >15 μm. The 10 classes of HC emissions data are: methane, paraffins/alkenes, olefins/alkenes, aromatics, carbonyls, oxygenated HCs, mercaptans (sulphur-containing compounds), halogenated aliphatics, other, and unidentified.

Other than the activities mentioned earlier, very little work has been done with respect to the temporal resolution of emissions data (i.e., quarterly). Data are limited to only a few sectors, and the present estimates are extremely crude. The major source of information for apportioning these data is quarterly fuel consumption statistics. Data for such activities as monthly sulphur or material balances and power plant electrial generation are scarce.

Emissions are estimated by any of the following methods (ranked in order of decreasing use):

- Standard emission factors on a sector basis, applied with plant-specific information;
- Consultants' reports, questionnaires, and other governmental data;
- Plant-specific emission factors used with other plant data;
- Material balances; and
- Source testing.

On a year-to-year basis, this ranking changes, but one can assume that the above list is generally correct.

Emission factors, the major tool used in developing emission estimates, are obtained from a number of sources: the U.S. Environmental Protection Agency's Compilation of Air Pollutant Emission Factors (Report AP-42), internally developed factors, company-specific factors, and other available literature.

Data for both controlled and uncontrolled emissions are reported. Data on pollution control equipment and equipment efficiency are available for some plants. Several bytes of computer capacity are made available in the NEIS per plant for storing these data along with either controlled or uncontrolled emission factors.

Hydrocarbon and VOC emissions are generally estimated by using emission factors (from AP-42 and other literature) and plant production statistics. In the case of petroleum refineries, data are available on process-specific operations from questionnaires. The major problem with using these data occurs when updating; emission factors must then be back calculated in order to be used with updated process flows.

A copy of the detailed record formats for point sources is attached.

AREA SOURCE DATA

The area source file is composed of hierarchical records containing data from nonfixed sources such as automobiles, fuel combustion, forest fires, etc.

Raw Data

As with point sources, the area sources (sectors) contributing the most pollution to the total burden vary according to the contaminant. The following area sources were the major polluters based on the 1978 inventory:

- Particluate matter--forest fires, slash burning, and industrial fuel combustion;
- SO_2--industrial fuel combustion, commercial fuel combustion, and residential fuel combustion;
- NO_x--gasoline-powered motor vehicles, diesel-powered engines, and industrial fuel combustion;
- HCs--gasoline-powered motor vehicles, gasoline and diesel marketing, and application of surface coatings; and
- CO--gasoline-powered motor vehicles, diesel-powered engines, and forest fires.

Transportation sources include the following: gasoline-powered motor vehicles (cars, motorcycles, snowmobiles, and light-, medium-, and heavy-duty trucks), railroads, aircraft, marine vehicles, offroad use of gasoline (agricultural equipment, heavy-duty construction, and industrial engines), diesel-powered engines (heavy-duty, agricultural, construction, and other diesel vehicles), and tire wear.

The types of raw data collected include: fuel consumption statistics, vehicle registrations, ships' calls in ports, landing/takeoff cycles for aircraft, refuse burned in commercial and industrial establishments, acres of forest burned, quantity of slash burned, fertilizer and pesticide application, solvent consumption, number of structural fires, and tobacco consumption. These data are usually available on a national or provincial basis. Depending on the base quantity, raw data are available either annually, seasonally (quarterly), or monthly. For example, fuel consumption statistics are quarterly, whereas data on forest acreage burned is annual. As with point source data, area source data are available on the NEIS for 1974, 1976, and 1978; additional information is available in hard copy for 1970 and 1972.

Emissions Data

The same contaminants discussed in the section on point sources apply in the discussion of area sources. Other than annual data, very little emissions information is available. Some statistics other than annual data are available, but the data have not been correlated. A first approximation of seasonal emissions would be the allocation of annual emissions evenly (25%) over all four seasons.

Area source emissions data can be resolved spatially in any of the following manners: national, provincial, census division, 127 km x 127 km grid, and census metropolitan area (CMA) for the 15 largest metropolitan areas in Canada. The starting point for apportioning area source data is the province. In order to obtain emissions on any other spatial resolution, a method of proration is used. The following parameters are used to prorate the provincial emissions: population, land area, number of dwelling units, labor force, acres of fertilized land, plant capacity, number of employees, landing/takeoff cycles for aircraft, and ship departures. These data are stored in proration tables in the NEIS for each year for which data are available. Emissions can then be determined by using the simple general equation:

$$\frac{\text{Parameter Value for Resolved Area}}{\text{Parameter Value for Province}} \times \text{Provincial Emissions} = \begin{array}{c}\text{Resolved}\\ \text{Area}\\ \text{Emissions}\end{array}$$

This standard technique is not in published form, although the argument values of the proration parameters are accessible through an NEIS printout.

Provincial emissions generated for area source sectors are calculated by using emission factors and base quantity statistics. In a few cases, control efficiencies are applied to uncontrolled emission factors (e.g., particulate emissions from coal combustion. Emission factors are mostly taken from EPA Report AP-42, but they can also be taken from other literature or derived internally. The cast approach is used widely for motor vehicle emissions, where emission factors are calculated by using actual data from domestically tested vehicles and emission standards applicable solely to Canada.

Emissions for VOCs are estimated by using two major references, EPA Report AP-42 and an internal report on the sources and emissions of VOCs in Canada. The latter document was not cited in the discussion of point sources, because it was not generally used as a reference for estimating point source emissions.

Whether they are total VOC emissions or emissions by compound class, all VOC area source emissions are calculated by using emission factors.

A copy of the detailed record formats for area sources is attached.

GENERAL INFORMATION

As discussed in the introduction, accurate data on the quantity and characteristics of emissions from all sources contributing to air pollution are required to assess the problem. Thus, an attempt has been made to incorporate as many sources of air pollution into the inventory as possible. Based on experience in developing the inventory over the past few years, data are complete to the extent possible. There are individual gaps in the plant file for many point sources, (e.g., stack data), but data have generally been sufficient for retrieval requirements.

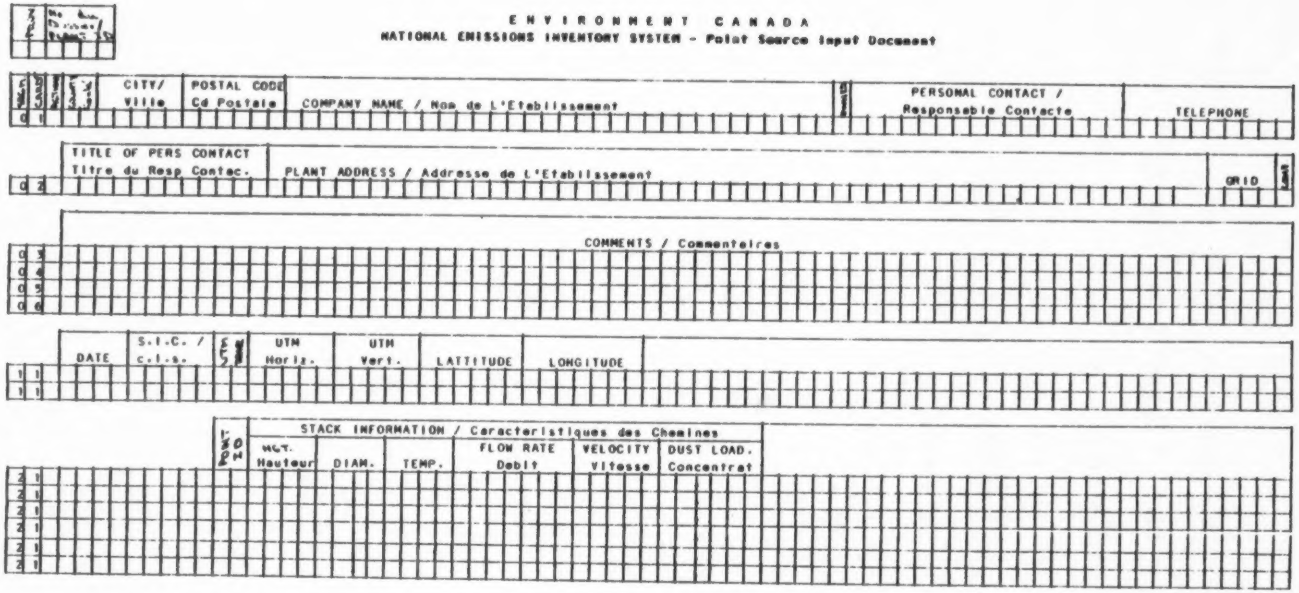
The most recent data on file is the 1978 inventory; data on major point sources have been updated to 1980. Present plans call for completion of the 1980 update within a year. Ideally, inventories should be as up-to-date as possible. However, due to the actual time required to assemble and complete data for an inventory, there is going to be a considerable lag in the completed product. For example, Statistics Canada, a major source of production and fuel consumption statistics, does not publish its results for 12 to 18 mo after the calendar year. Thus, 1980 statistics are usually not available until mid-1982.

There is no formalized quality assurance program for the emissions reporting developed in the inventory. Information on a particular plant may be obtained from multiple sources (e.g., provincial agency, company questionnaire), and there is always the possibility that emissions calculated from these sources may be significantly different. It is then the project engineer's responsibility to use his/her expertise and judgment to determine which source is correct. This can be achieved in different ways (e.g., direct contact with company, personal experience in that industry sector).

Estimates of the overall precision of the inventory have been made for two contaminants, SO_2 and NO_x. A critical review of the methodologies used to determine information sources was made prior to establishing which factors determine the confidence level of the inventory. Spot checks and systematic reviews of emissions versus production and/or previous emission rates were also made. The results of this exercise showed that the precisions of the two inventories were ±6.3% and ±10.3% for SO_2 and NO_x, respectively.

Maintaining the quality of the data is the responsibility of the Inventory Management Division. Random checks of the data prior to and following input to the NEIS are necessary in order to ensure that emissions data are reasonable relative to historical information. Both an internal and external review of inventory data is necessary to maintain the quality of the data. When a draft report is completed, it is forwarded to Federal regional and provincial offices for technical comment, as well as to other divisions within Environment Canada, to ensure that data have been used correctly. Both summarized reports and individual plant files (in computer format) are forwarded. Reviewer comments are analyzed and, if necessary, the report is revised.

Presently, data (point and area sources) are manually input to the NEIS through the use of updated files. Manual entry necessitates strict control of the inputs from the original keypunching. Once it is ensured that all inputted data are correct, retrievals on any level are possible, for any parameters required. For example, plant emissions can be determined by stack or summed for the entire plant, or total emissions can be retrieved by sector for each province. There is great flexibility in requesting retrievals at any level needed.



ENVIRONMENT CANADA
NATIONAL EMISSIONS INVENTORY SYSTEM - Point Source Input Document

0 1 CITY/ Ville | POSTAL CODE Cd Postale | COMPANY NAME / Nom de L'Etablissement | PERSONAL CONTACT / Responsable Contacte | TELEPHONE

0 2 TITLE OF PERS CONTACT Titre du Resp Contac. | PLANT ADDRESS / Addresse de L'Etablissement | GRID

COMMENTS / Commentaires
0 3
0 4
0 5
0 6

1 1 DATE | S.I.C. / c.i.s. | UTM Horiz. | UTM Vert. | LATTITUDE | LONGITUDE
1 1

STACK INFORMATION / Caracteristiques des Chemines
HGT. Hauteur | DIAM. | TEMP. | FLOW RATE Debit | VELOCITY Vitesse | DUST LOAD. Concentrat
2 1
2 1
2 1
2 1
2 1
2 1

Figure 1. National Emissions Inventory System point source input document (Environment Canada).

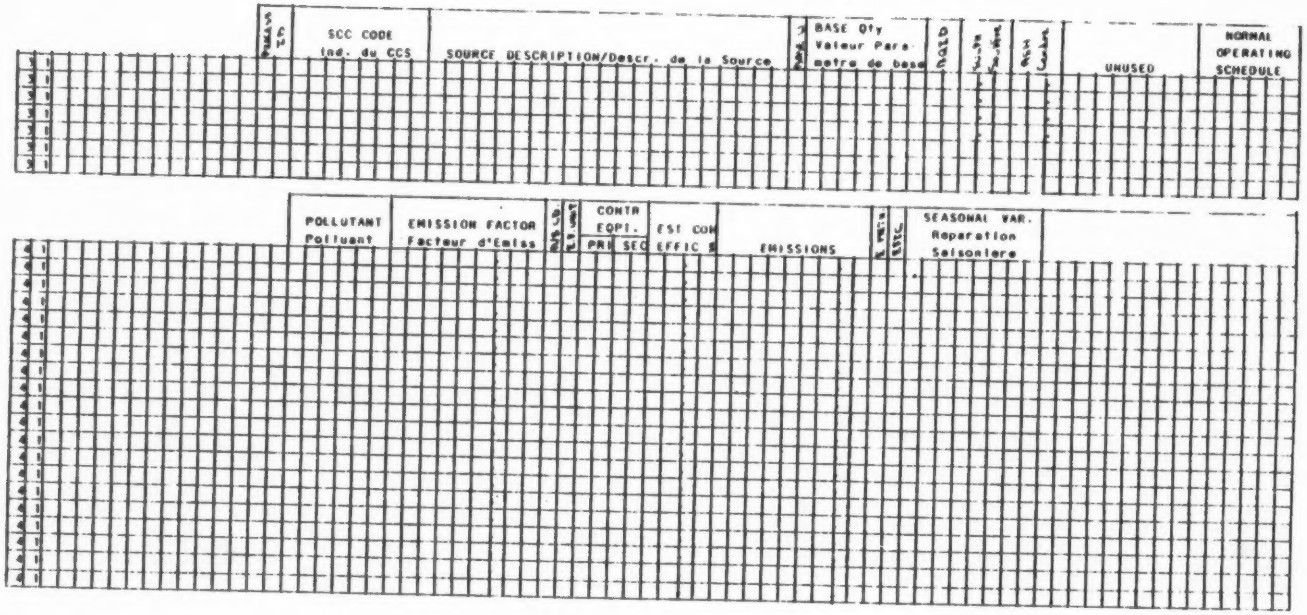

SCC CODE
Ind. du CCS
SOURCE DESCRIPTION/Descr. de la Source
BASE Qty
Valeur Para-
metre de base
UNUSED
NORMAL
OPERATING
SCHEDULE
POLLUTANT
Polluant
EMISSION FACTOR
Facteur d'Emiss
CONTR
EQPT.
PRI SEC
EST CON
EFFIC %
EMISSIONS
SEASONAL VAR.
Reparation
Saisoniere

Figure 2. National Emissions Inventory System sample form.

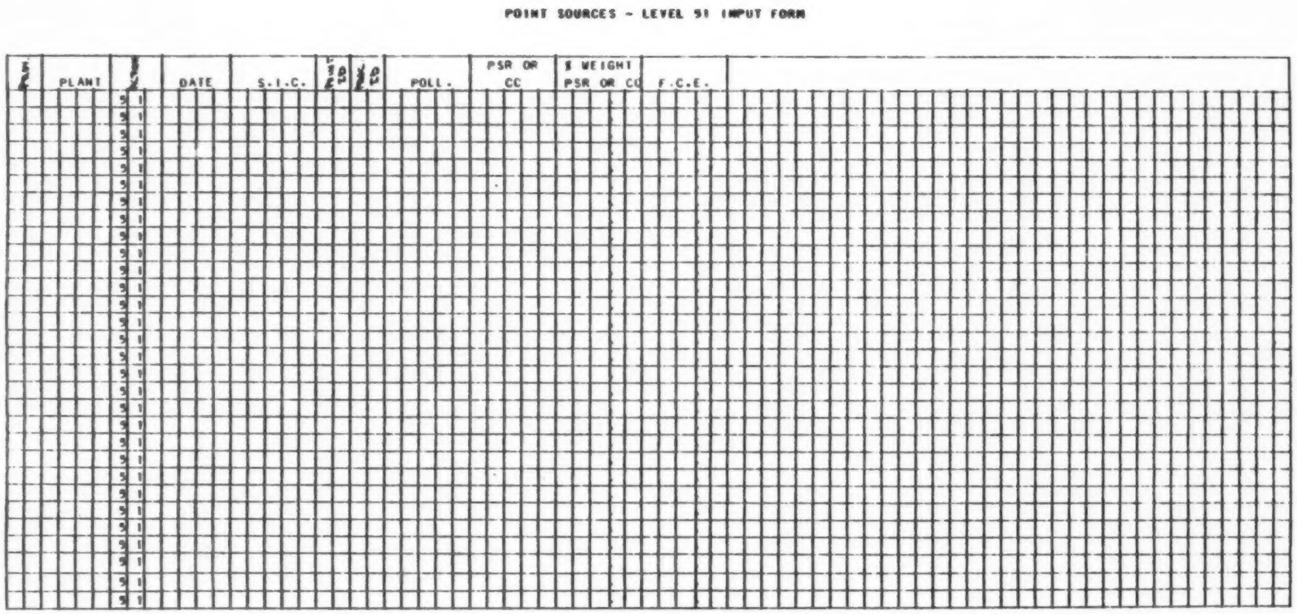
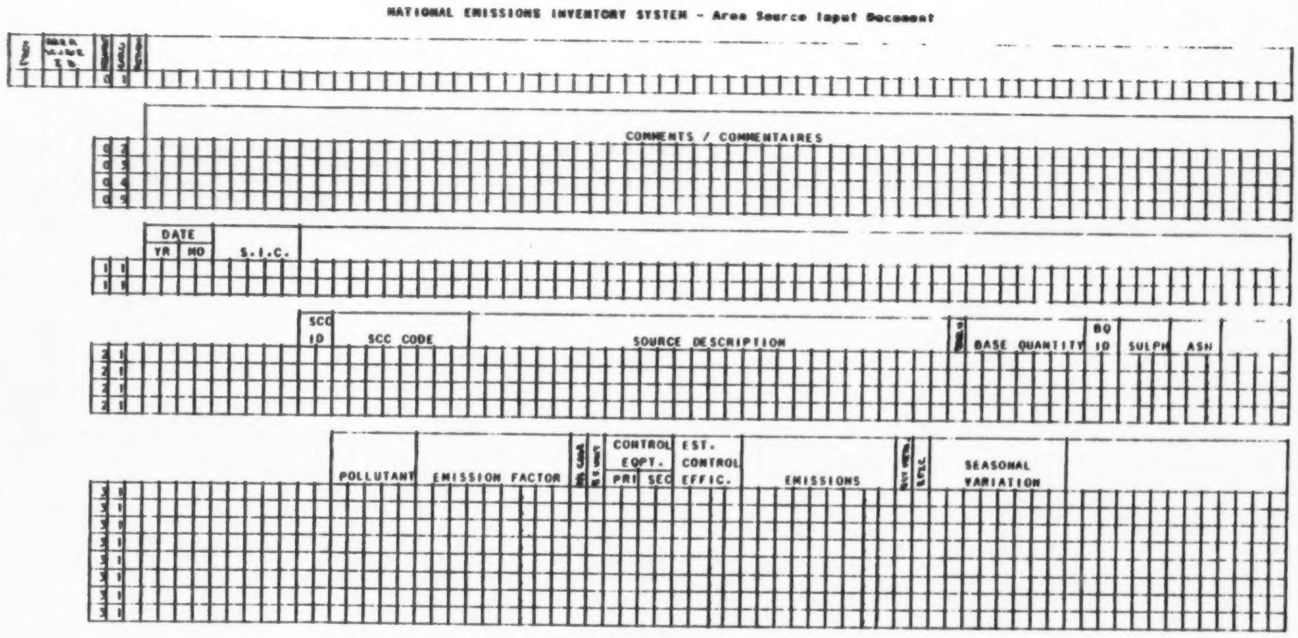

POINT SOURCES - LEVEL 51 INPUT FORM
PLANT
DATE
S.I.C.
POLL.
PSR OR CC
% WEIGHT PSR OR CC
F.C.E.
5
1

Figure 3. National Emissions Inventory System point sources, level 51, input form.



NATIONAL EMISSIONS INVENTORY SYSTEM - Area Source Input Document

COMMENTS / COMMENTAIRES

DATE

YR

MO

S.I.C.

SCC ID

SCC CODE

SOURCE DESCRIPTION

BASE QUANTITY

BQ ID

SULPH

ASH

POLLUTANT

EMISSION FACTOR

CONTROL EQPT.

PRI

SEC

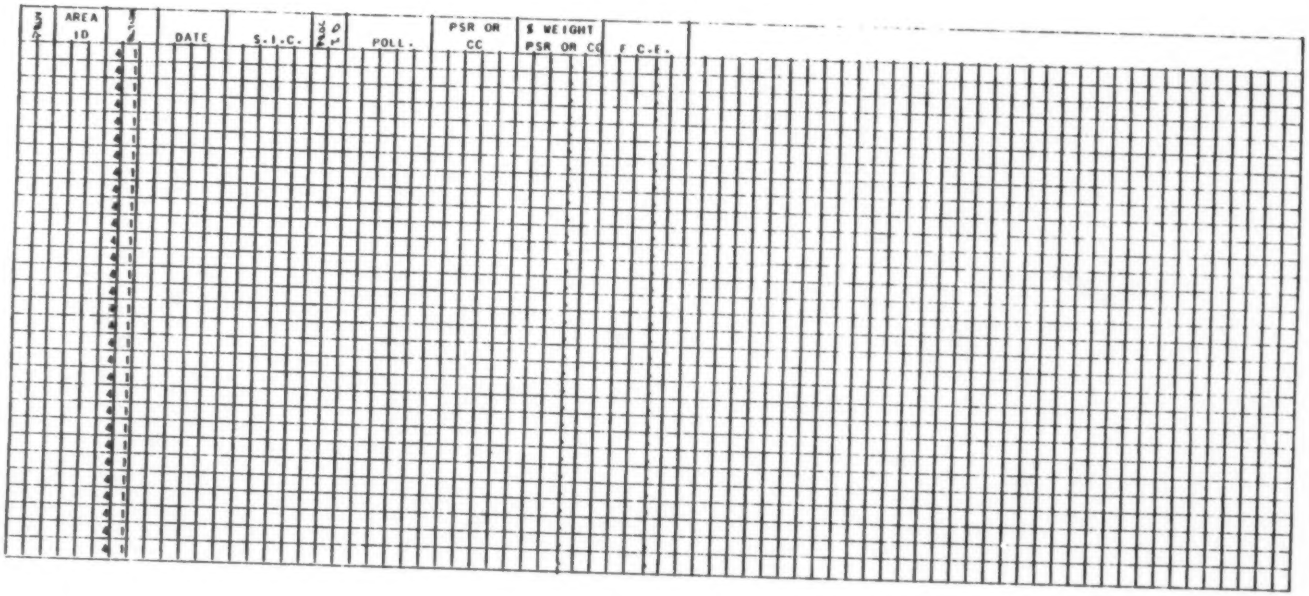
EST. CONTROL EFFIC.

EMISSIONS

SEASONAL VARIATION

Figure 4. National Emissions Inventory System area source input document.

AREA SOURCES - LEVEL 41 INPUT FORM



AREA ID
DATE
S.I.C.
POLL.
PSR OR CC
$ WEIGHT PSR OR CC
F C.F.
4 1

Figure 5. National Emissions Inventory System level 41 input form.

OECD PRESENTATION*

Anton Eliassen

Norwegian Meteorological Institute
Blindern, Oslo 3, Norway

Although I am not an emissions expert, I have been using emissions data for some time. It is therefore perhaps easier for me than the emissions experts to say that not everything is fine in the field of emissions inventories.

I work at the Norwegian Meteorological Institute, which is one of two institutes responsible for meteorological dispersion modeling in the European program on the long-range transport of air pollutants. This is a United Nations program, which operates under the United Nations Economic Commission for Europe (ECE). It is also part of the convention we have in Europe on long-range transboundary air pollution that has just entered into force.

This program is now an activity under the convention, where it is referred to as EMEP. Basically, we have been studying sulfur transport up to now, but we expect that since the convention has entered into force, we will include other species.

To start with the sulfur emissions, these are given in a report here, which is available upon request from the Institute. This is 1978 emissions data for SO_2 in Europe. These are data from both the Eastern and Western European countries. I think the emissions from Eastern Europe are bound to be relevant in the oxidant problem because, whenever there is a high pressure area with good chances of oxidant formation over Europe, you will have mostly easterly winds and you will have transport from the Eastern countries towards the west, with time for oxidant production along the way.

The problem is that Europe consists of several independent countries, each of which decides what sort of data to provide for this work. Up to now, we have received at least one number for sulfur emission from each country. However, some of the Western European countries have given us fairly detailed emissions inventories for SO_2. So, this is the information we have to work with.

This is an example of an emissions grid for Czechoslovakia. We received one number from Czechoslovakia, which was submitted to ECE. However, we know the location and type of industries found there, and it is amazing how much information it is possible to find. We know the locations of the cities and the populations of the cities. So, we try to distribute as best as we can this total number over Czechoslovakia. This is how we work.

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Other countries, for example the Federal Republic of Germany, have provided very detailed data. In such cases, we obviously get much better information. However, there is no point in using such detailed information from one country when we may have only one number for several other countries. Therefore, we grid the data from the Federal Republic of Germany onto the 150-km emissions grid that we are using to run the model. So, the trouble is the vast difference in the quality of data submitted by the different countries.

On the map shown here, which illustrates total SO_2 emissions, you do not see numbers but colors to indicate the most important emissions squares. The highest emission in the whole grid is in the Soviet Union. In face, we have some additional data for that particular area. The only other thing we have is a total emission number for SO_2, which is for the whole country, including Siberia. It is quite difficult to grid that information in.

This is the situation that we have to work with. We hope that it will improve, but it appears to be a quite sensitive area.

As for NO_x, this is basically the data that we used to perform the model calculations Dr. Hov discussed. The NO_x emissions data are available in this report from the Norwegian Institute for Air Research.

I will try to outline briefly how these numbers were estimated. For OECD-Europe, we have fairly good data, based on emission factors and fuel consumption. From there, we obtained national NO_x emission estimates, which are available from OECD reports.

As for Eastern Europe, we obtained one number, an annual NO_x emissions number for each country in the OECD region. Then, we relate that number to energy consumption, using energy consumption data that were available from the United Nations Statistical Office. That included data for both Eastern and Western European countries.

If you look at the data for OECD-Europe, you can relate energy consumption on the horizontal axis to the NO_x emissions on the vertical axis. First, you have to remove the energy consumption that obviously does not produce NO_x. After that, you get quite a nice relation. So, we think that the best thing to do for Eastern Europe is to take the energy consumption data from the United Nations Statistical Office and to use this same relation to estimate their NO_x emissions.

We needed these data in the grid to perform calculations with the model. We distributed the NO_x emissions according to the SO_2 emissions that we had estimated earlier, with a few exceptions.

Then comes the most difficult part, the HCs. From OECD, we again have some estimates, NMHCs, and we use those that the different countries have submitted to OECD. Of course, we do not always know the basis for those numbers.

The difference in the ratio of hydrocarbon-to-nitrogen emissions among OECD European countries varies between 0.5 and 1.8. The only thing that we can possibly do at present for Eastern Europe is to assume a ratio of 1 for these emissions. Perhaps it is a little too low.

To summarize, I can present a table showing the national estmated emissions of the species I have talked about. If we get any complaints for countries who disagree with these estimates, we assume that we were wrong and ask for suggestions. For example, in the case of Romania, we were informed that we should divide our sulfur emission estimate by 10.

These data form the basis of any model calculation to include all of Europe. The experience is that, even if you have countries with very good emissions data and you try to run sophisticated models within these countries, many of the calculated values are determined by the flux across the air model boundary. In such cases, it is very difficult to avoid the problem of data availability. So, I thought that I would point out this problem, so we do not forget it under our next discussions. I think this also shows that there is no point in using an extremely sophisticated model with very bad emissions data.

OECD PRESENTATION*

Peter Builtjes

MT-TNO, Department of Fluid Mechanics
The Netherlands

The emissions inventory in The Netherlands was made by TNO by order of the Ministry of the Environment. It contains air pollution data as well as water pollution data. The first phase started about 1974 and it is about complete now. We are now in the second phase of updating.

The last year for which emissions data are available is 1980. The information for large industries was obtained by inspection and the information for small industries was obtained by questionnaire. Inspection can, of course, be easily done for a small country like The Netherlands.

The issue of confidentiality presents some limitations for real numbers for the industry, but there are old tapes on file somewhere. These are updated yearly by questionnaire for large industries. For other industries, these are updated every 3 yr.

Something must be said about point sources, stack information, location, and stack height, temperature, etc. Otherwise, we cannot use the information for dispersion calculations. Area sources are divided into 1 x 1 km grids, which include things like heating. The species are NO_x, equivalent NO_2, SO_2, CO, and HCs. There is also some information about things like heavy metals, particulates, and ammonia.

As to accuracy, particularly with regard to total NO_x emissions in The Netherlands, the guess is that it is accurate within 10%. For HCs, it is accurate within 30%.

There are other things like annual average values and information for traffic, days, etc, to be considered. All of this information is available on file or on tape, so we can use it, as we did for the chemical studies. We can process it and we can directly use the information connected with our dispersion model.

Let's say something about scenarios. When we do a real control strategy, we use scenarios and abatement reference cases. We use abatement reference cases to say we could put down this industry or have this requirement for traffic. There is also a scenario system, which means you can assume a certain gross national product and then allocate in a balanced way which industry could increase and which industry could decrease.

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I will conclude my remarks by showing you two slides. The first slide shows NO_x emissions for 1980 in The Netherlands. Because, The Netherlands is so small, you cannot do any real calculations for the surrounding countries. So, we also used information from Germany.

You can clearly see the area including the Rhine, the Ruhr, and Antwerp. This is an area of approximately 305 km^2. These are grids of 10 km x 10 km. This is just for the NO values of the total NO_x values for 1977, and specifically just for mobile sources, traffic, for The Netherlands. Road traffic and auto traffic, 270 stationary sources--power stations, industry, we interpreted around 500 x 10^6 kg.

For the Federal Republic of Germany, it is only about 1,500 kg and for Belgium and Northern France it is about 370 kg.

DISCUSSION FOR OECD PAPERS

D. Jost: Towards the end of the year, we will have available a nationwide emissions inventory for NO_x, which is based on energy consumption and energy production. It will be divided into grid sizes in the range of 20 km, and it will be based on data from villages.

B. Luebkert: Will that only be available on a grid basis or will it also be available with respect to source categories?

D. Jost: The original data will be available on a grid base of 1 km x 1 km. Within that framework, one is free to locate the known point source. We know there are point sources within this base, but we do not give the exact locations.

B. Luebkert: That is not quite what I mean. Will it give information all together on a nationwide basis? Will it classify how much is emitted by each industry source category?

D. Jost: Yes.

B. Luebkert: So, you have one figure for traffic and you have one figure for the chemical industry. Will that be available too?

D. Jost: This will be available. It will not be printed out at once, but it could be obtained from the system.

H. van Dop: Will there also be an indication of source height in that emissions inventory?

D. Jost: There will be source height categories, although not exactly for each source height. I do not know the numbers, but there will be several categories within this system, which we thought would be good enough for calculating medium-range transport. I cannot give you the area at this moment.

L. Kropp: To add to that, from the emissions declarations that have to be given by large-power-plant operators, we will be able to get all of these source heights for the individual sources of the heavily polluted areas.

GENERAL DISCUSSION FOLLOWING SESSION II

Lars Lindau, Chairman

L. Lindau: I would like to review some questions that I posed this morning. The new point is, from the discussion in OECD, whether we are going to have a project on total chemical oxidant modeling and emissions inventories. What are the main difficulties and where are the data errors when you are establishing inventories? You could list a lot of different questions from that.

In addition, no one has talked about emissions from vegetation and forests and how they have to be dealt with.

There have been some discussions about currentness, time variation, confidentiality, things like that.

Some of these questions have already been brought up during the discussion, but how big are the errors in making emission estimates? You have to know whether these are 10%, 30%, as was talked about from the Dutch side, but perhaps they are much more. Are they of a factor of 2 or a factor of 5? How big are the errors?

What does it mean to the output of a model if there are large error variations in emissions data from different areas? That important question was raised here before by Anton Eliassen, especially for the European case where you have trouble getting data from the Eastern countries.

Is it worthwhile to raise emissions data from the Western countries?

What is the cost of establishing an emissions inventory for NO_x and different process HCs? For example, would a grid size of 100 km, some sort of area could be Germany or some other area, and some sort of error, some sort of reliability, and then some sort of breakdown of the emissions.

Another question that is important is: Which error in emissions data can you accept when you are discussing the end result, the control strategy? What is the need for reliability in the emissions data we are using when we are using the models for control strategy purposes?

H. van Dop: Anton Eliassen mentioned an interesting thing, that it might be possible to construct emissions inventories in an indirect way. This obviously requires industries and so on, but it can be done by just making smart guesses.

It would be useful to compare these guesses with emissions inventories, with well-known emissions inventories, to prove if this method can be used anywhere you want. I would like to ask Anton if that has been done. Have you compared your guesses with existing emissions data?

A. Eliassen: Unfortunately we did not think of that. For countries supplying data that we thought were accurate, we used those. We did not try to make so-called intelligent guesses.

L. Kropp: In this context, I compared the data you just mentioned in the first and second slide (between) the NO_x emissions of European countries presented in the paper. I realized that these data were about 30% less than the data I found for different emissions inventories. I first thought that NO_x was given as NO.

J. Bosch: Our responsibility is the National Emission Data System and national estimates. We have two means of estimating emissions. One is a "bottom-up" approach whereby we obtain individual plant-by-plant data nationwide. The other is a "top-down" approach whereby we use Department of Commerce and national fuel consumption figures.

Actually, in comparing the two of them, the difference in variance is less than 5%, generally about 2%. So, this is a rather independent means of estimation that could actually be applied to any nation that is obtaining national data on fuel consumption, which is quite common.

E. Runca: I would like to make a general comment on model application. Considering the difficulties in obtaining emissions data in Europe, models must be adequate to the data available. This is true if you want to apply the model to the whole of Europe, including Eastern and Western counties.

On the other hand, for other regions of Europe, it might also be interesting to have a monitored data base, monitored data, and to apply more sophisticated models to verify that some assumptions are valid and some strategies are correct. So, I do not think that we have only to look at the general description of the problem in Europe, including Eastern and Western countries.

G. Whitten: As a modeler and a user of some of these emissions data, a powerful cross-check amongst emissions inventories is a per-capita emissions of HCs, a per-capita emissions of NO_x, and perhaps an HC-to-NO_x ratio determined on a large scale. One might subtract the power plant emissions, since they are a large elevated source of NO_x. Sometimes, when you go to a per-capita emission of HCs based on total fuel consumption, you lose some of the range in the HC-NO_x ratio that you see in emissions inventories and you also provide a cross-check within the inventory itself.

Another procedure that we have used is to look at the overall speciation. An important aspect at any emissions inventory is the emission of carbonyl compounds like aldehydes. Occasionally, they get left out, and the models are sometimes very responsive to these things. Also, you can look at a cross-check between other emissions inventories, a very useful procedure.

SESSION III

AVAILABLE AEROMETRIC DATA BASES

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AVAILABILITY OF OZONE AND OZONE PRECURSOR DATA FROM THE SAROAD SYSTEM*

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INTRODUCTION

The Storage and Retrieval of Aerometric Data (SAROAD) system was developed in 1966 by the Federal Government when it became apparent that the large volume of data generated by ambient air quality monitoring required management by a computerized system. The volume of data used to establish SAROAD initially involved a network of approximately 300 sites. This network, the National Air Sampling Network, began operation in 1957, generating approximately 100,000 pollutant volumes nationwide a year.

SAROAD has evolved from this initial system to a complex system that currently stores over 200 million pollutant values collected from over 16,000 sites operated by Federal, state, and local agencies. The 4,000 to 5,000 active sites in SAROAD report approximately 20 million data values annually. The remaining 11,000 to 12,000 sites are no longer active, but they are retained for historic information.

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*This paper has been reviewed by the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

Growth in the volume of data collected has mandated standardized procedures for site characterization, sampling method identification, data editing and validation, and data analysis and summarization. In order to use these data for analysis, over 30 programs have been standardized and are available for retrieval and display of both raw data and summary statistics.

DATA COLLECTION AND REPORTING REQUIREMENTS

The change from a small Federal sampling network to a large nationwide network operated by Federal as well as state and local agencies was a result of the 1970 Clean Air Act Amendments. This legislation, which established the U.S. Environmental Protection Agency (EPA), required EPA to develop and implement National Ambient Air Quality Standards (NAAQS) for six pollutants: TSP, SO_2, CO, O_3, HC, and NO_2. (The NAAQS for HC was recently rescinded because it is used only as a guide in devising control programs for O_3.)

The Clean Air Act Amendments required that primary ambient air quality standards, designed to protect the public health, be met nationally by 1975 unless a 2-yr extension was granted by the EPA Administrator. Secondary standards, designed to protect the public welfare, had to be achieved within a reasonable time. Each state was required to develop and submit to EPA a State Implementation Plan (SIP) to identify corrective actions to be taken to reduce air pollution to meet the NAAQS.

Each SIP was to include the design and operation of an ambient air monitoring network to determine compliance with the NAAQS. In addition to operating the monitoring network and collecting ambient air quality data, state and local agencies were required to report the concentrations to SAROAD each quarter, with 45 days after the end of the quarter. Historic data collected and utilized to develop the SIP were also to be reported. These data were to be used to:

- Judge compliance with and/or progress made toward meeting ambient air quality standards;
- Activate emergency control procedures to prevent or alleviate air pollution episodes;
- Observe pollution trends; and
- Provide a data base for evaluating the effects of urbanization, land use, and/or transportation planning; for developing and evaluating abatement strategies; and for developing and validating diffusion models.

When the NAAQS were promulgated by EPA in 1971, standard monitoring methods (reference methods) were also promulgated for each pollutant. These reference methods were the methods most often used by EPA to collect ambient concentrations and were thought to be the most reliable. As agencies began routinely utilizing the reference methods, methodological problems were encountered. Also, differences were noted in the sample collection time, sampling procedures, and sampling instrumentation required for the NAAQS. Automated instrumentation was developed to sample for criteria pollutants, which created in turn the problem of comparing sample results from different chemical analysis procedures.

These problems were resolved in 1975 when EPA regulations established an "equivalency" method between reference methods and "candidate" methods. The regulations prohibited state and local agencies from purchasing any new instrumentation that was not equivalent and permitted the utilization of existing nonequivalent instrumentation until February 1980. The equivalency regulations identified test procedures and reporting requirements that each instrument manufacturer must complete to allow EPA to determine whether the candidate method was equivalent. The manufacturer was also required to supply an instrument manual to the purchaser to identify mandatory operating parameters.

Table 1 lists criteria pollutants and the reference methods and number of equivalent instruments available for each. Table 1 does not include HC because the HC standard is only used as a guide in developing the SIP to meet the O_3 standard. HC monitoring is not currently required but is performed by some state and local agencies.

Although the regulations for SIPs specified an approximate number of sites for a given geographical area, based on population, they did not specify location. This resulted in inconsistencies in site locations and specific probe locations, such as height above ground and distance from the nearest street or road. These variables have been studied, and specific guidelines have since been developed and implemented to standardize these parameters.

Specific quality assurance procedures were not specified as part of the initial SIP regulations. Quality assurance procedures were left up to the individual state and local agencies to develop and implement.

TABLE 1. CRITERIA POLLUTANTS AND MEASUREMENT METHODS

Pollutant	Reference Method	Number of Reference Equivalent Instruments
Suspended particulate matter	High-volume	None
Pb	High-volume/atomic absorption	4
SO_2	Pararosaniline	15[a]
CO	Nondispersive infrared	9[b]
O_3	Chemiluminescence	14[c]
NO_2	Chemiluminescence	13[d]

[a] Two equivalent procedures for analyzing samples collected by the reference method and 13 equivalent instruments sampling continuously.
[b] All instruments use the reference method measurement principle.
[c] Nine instruments use the reference method measurement principle.
[d] Ten instruments use the reference method measurement principle; three use a manual sampling technique and laboratory analysis.

In 1975, the Quality Assurance and Environmental Monitoring Laboratory of the EPA Office of Research and Development (ORD) began compiling quality assurance manuals. These manuals consolidated previous guidelines into a centralized source of quality assurance information. Volume I of the manual defined the quality assurance function in the air pollution control agency and identified the procedures required for training, preventive maintenance, sample collection, sample analysis, data reporting, calibration, audit procedures, data validation, etc. Volume II defined the quality assurance procedures for the specific reference and equivalent methods for each criteria pollutant: suspended particulate, NO_2, SO_2, CO, O_3, and Pb.

NAMS/SLAMS REPORTING REQUIREMENTS

In 1979, as a result of deficiencies in state and local agency air quality monitoring and data reporting programs and of revisions to the 1977 Clean Air Act Amendments, EPA promulgated new regulations that changed the air quality data collection and reporting requirements. These changes were made to improve the timeliness and quality of air monitoring data. The revised regulations identified the National Air Monitoring Stations (NAMS) and the State and Local Air Monitoring Stations (SLAMS).

NAMS is a limited network of approximately 1,200 stations required by EPA to perform national data analysis. In this network, sites are located in large urban areas, with the objective of measuring ambient concentrations in areas of high pollutant concentrations and/or high population exposure. These sites monitor for gaseous pollutants, using only continuous instruments. The raw data values are submitted quarterly, within 90 days after the end of the quarter.

SLAMS is an expanded network of approximately 5,000 sites required by individual states to determine violations of NAAQS. This network was designed by the states primarily to meet their needs. The size is based on such factors as meteorology, geography, population, and emission density. The NAMS network is a subset of the SLAMS network. For SLAMS, the states are required to submit an annual summary, within 6 mo after the end of the year.

The regulations promulgated in 1979 required that the NAMS network be established and operational by January 1, 1981, and that the SLAMS network be established and operational by January 1, 1983. The regulations involved the following additional specifications for both NAMS and SLAMS:

- A quality assurance plan by state and local agencies, approved by EPA--Precision and accuracy information must be collected and reported to assess the sampling and analysis procedures. The agency must review and evaluate this information to identify possible problem areas and to initiate corrective action such as more frequent calibration or maintenance.
- Ambient monitoring methodology--For the NAMS network, only reference or equivalent methods that monitor continuously were permitted for gases and only the reference method was permitted for suspended particulates. For the SLAMS network, only reference or equivalent methods were preferred, but procedures were defined to permit states to obtain limited approval of other methods.
- Network designs for SLAMS and NAMS--Design criteria included parameters such as emission sources, meteorology, and geography to evaluate the existing sites. Other parameters, such as the different measurement scales (micro, middle, neighborhood, urban, or regional) that are appropriate for SLAMS and NAMS and the number of NAMS sites that are required, based on population and approximate concentration ranges, were also defined.
- Probe siting criteria--The material used to make the probe should be nonreactive. In addition, a maximum residence time for the sample in the probe was established, and restrictions on the probe location at the sampling site were made, including vertical and horizontal probe placement, distance from obstructions, and distance from roads as a function of traffic.

NAMS/SLAMS reporting requirements have been implemented as scheduled to provide air monitoring data of acceptable quality, comparable data from all monitoring stations, and timely data submission for national assessment purposes.

THE SAROAD SYSTEM

As previously discussed, SAROAD was intially developed to store small volumes of data. As requirements were implemented for collecting and reporting air quality data by state and local agencies, the volume of data acquired mandated the development of uniform coding procedures and formats, codes to identify the data, and procedures to edit, validate, summarize, and report the data.

SAROAD is operated by the National Air Data Branch (NADB). The organizations supplying air quality data to SAROAD include state and local agencies and the 10 EPA Regional Offices. These organizations are responsible for the following functions:

- State or local agencies establish the monitoring sites, operate the equipment to sample the ambient air, convert the ambient air data to a format compatible for storage in SAROAD, and submit these data to their respective EPA Regional Office.
- The EPA Regional Office approves the sampling sites established by the state or local agency; edits, validates, and corrects data with assistance from the submitting agency; submits the data to NADB for update; and returns data for standards violation and trends in air quality.
- NADB manages SAROAD by updating data obtained from the EPA Regional Offices; by developing and maintaining SAROAD software necessary to edit, store, and analyze the data; by developing procedures and codes for processing data; and by providing data to users when requested.

Before air quality data from a site can be stored in SAROAD, the site must be registered. This involves assigning an identifier to the site and using the identifier when any data are reported. The identifier includes the state and city or county where the site is located and the site number within the city or county. Other data items that are stored for the site include: the geographical coordinates, the time zone, the Standard Metropolitan Statistical Area (SMSA) code, the agency name, the site address, the station type (center city-commercial), the elevation above ground and above sea level, the Air Quality Control Region (AQCR) code, the site type (NAMS or SLAMS), and details describing the site. The population of the city and AQCR are also included.

SAROAD permits the storage of ambient concentrations for any pollutant or parameter that can be defined and measured with an acceptable analytical procedure. Each pollutant is assigned a code, and each different sampling and analysis procedure is assigned a method code. Other codes that are assigned include the units code, which identifies the units of measure, and the interval code, which identifies the sampling frequency. The sampling frequency can vary from long-term monthly exposures to short-term continuous sampling reported as hourly averages. The complete identifier for a data value includes the site identifier; the year, month, day, and hour; and the pollutant-method-interval-unit codes.

In addition to storing site information and raw data, SAROAD computes and stores summary statistics on a quarterly and annual basis. These summary statistics are site- and pollutant-specific and include statistics such as arithmetic and geometric means, standard deviations, maximum and minimum values, violation counts for criteria pollutants, and the percentile distribution.

AMBIENT DATA AVAILABLE FOR TRANSPORT MODELS

As discussed in previous sections, state and local agencies are required to monitor and report ambient air quality concentrations to SAROAD for the NAAQS pollutants. In addition to these criteria pollutants, many state and local agencies collect and report data for several meteorological parameters. These data are not required but are collected and reported at sites monitoring for other pollutants.

Table 2 lists pollutant and meteorological data reported to SAROAD that might be of value for input to O_3 transport models. Although most state and local agencies began reporting data in 1971 or 1972, only data for the last 5 yr are summarized. The count represents the number of sites reporting data for each pollutant each year. The number of sites reporting data for NO_2 and O_3 is most consistent from year to year. The number of sites reporting data for the other pollutants and meteorological parameters peaked in 1980 and has now begun to decrease. The reduction in required monitoring data probably reflects the implementation of NAMS/SLAMS regulations that are decreasing the total resources available for monitoring.

In order to reduce the volume of data presented in Table 2, the third quarters for 1980 and 1981 were selected for additional analysis. These time periods were selected because the third quarter is the O_3 season for all states. The third-quarter data for 1982 were not utilized because the data were not complete for all states. Also, the data for 1980 and 1981 were selected because

TABLE 2. AVAILABILITY OF DATA BY POLLUTANT AND YEAR

Pollutant	Number of Sites for Each Year				
	1978	1979	1980	1981	1982
NO	200	183	219	175	138
NO_2	298	314	341	344	278
NO_x	114	153	185	164	144
THCs	126	103	88	60	36
NMHCs	83	76	86	64	28
CH_4	67	60	72	42	14
O_3	581	651	796	773	656
Wind speed	232	242	317	252	211
Wind direction	219	244	310	251	210
Lapse rate	0	0	0	0	0
Mixing height	0	2	1	0	0
Temperature	138	103	135	129	106
Temperature difference	4	12	13	12	11
Solar radiation	28	18	25	8	5

the quality of data is expected to be better than that for previous years as a result of implementing NAMS/SLAMS monitoring requirements.

Tables 3 and 4 present the number of sites reporting data to SAROAD by state for the pollutants listed in Table 2. The counts reflect the number of sites collecting and reporting not only O_3 data but also other general

TABLE 3. NUMBER OF SITES REPORTING DATA FOR THIRD QUARTER, 1980

	Type of Data Reported[a]					
State	O_3	O_3, NO_x	O_3, NO_x, HC	O_3, MET	O_3, MET, NO_x, HC	O_3, NO_x, HC, MET
Alabama	7			1	1	1
Alaska					1	
Arizona	8	1	1	5	1	
Arkansas	1	1				
California	68	35	34			
Colorado	5	2	1	1	1	
Connecticut	1			7	2	
Delaware	5	1				
District of Columbia		2				
Florida	11	1		7	3	
Georgia	5					
Hawaii	1					
Idaho						
Illinois	36	3		2		1
Indiana	12			1	1	
Iowa	3	2				
Kansas	3					
Kentucky	11	1			7	1
Louisiana	5					
Maine	6			1		
Maryland	1	1		8	11	
Massachusetts	7	1		3	7	
Michigan	14	3				
Minnesota	8		1			
Mississippi	2	1				
Missouri	5	1		1	4	7
Montana	2				2	
Nebraska	3	1				
Nevada	2		1			
New Hampshire	1			1		
New Jersey	5	7		1	5	
New Mexico	5	1				
New York	17	5		3	4	
North Carolina	11					
North Dakota					2	
Ohio	24	3	1	2	2	3
Oklahoma	1	5				

(continued)

TABLE 3. (continued)

State	Type of Data Reported[a]					
	O_3	O_3, NO_x	O_3, NO_x, HC	O_3, MET	O_3, MET, NO_x, HC	O_3, NO_x, HC, MET
Oregon	7					
Pennsylvania	9	3	1	2	2	16
Rhode Island	1			1		
South Carolina	6			2		
South Dakota						
Tennessee	6				1	
Texas	9		2	7	10	6
Utah		4				
Vermont	3					
Virginia	10	7			1	
Washington	8					
West Virginia	2	1				
Wisconsin	14	5	1			
Wyoming						
TOTAL	371	99	43	56	68	35

[a] Sites were counted as reporting NO_x if NO, NO_2, NO_x, or any combination was reported; HC if THCs, NMHCs, CH_4, or any combination was reported; and MET if any meteorological parameter from Table 2 was reported.

TABLE 4. NUMBER OF SITES REPORTING DATA FOR THIRD QUARTER, 1981

	Type of Data Reported[a]					
State	O_3	O_3, NO_x	O_3, NO_x, HC	O_3, MET	O_3, MET, NO_x, HC	O_3, NO_x, HC, MET
Alabama	8				2	1
Alaska					1	
Arizona	13	2			2	
Arkansas		2				
California	59	36	32			
Colorado	7	2	1		2	
Connecticut	1			4	6	1
Delaware	4					
District of Columbia		1	1			
Florida	8	3		9	1	
Georgia	4					
Hawaii	1					
Idaho						
Illinois	33	2		3		
Indiana	13			2	1	1
Iowa	4	1				
Kansas	2				1	
Kentucky	22	3			5	
Louisiana	12	1				
Maine	5			1		
Maryland	9	9				
Massachusetts	7	1		4		2
Michigan	13	3				
Minnesota	9		1			
Mississippi	3					
Missouri	2	2				
Montana	1		1			
Nebraska	4					
Nevada	5	1		5	4	2
New Hampshire	5				1	
New Jersey	7	7				
New Mexico	7	1	1			
New York	23	2				
North Carolina	11					
North Dakota		2				
Ohio	24	3	1	2	2	2
Oklahoma	4	3				

(continued)

TABLE 4. (continued)

State	Type of Data Reported[a] O_3	 O_3, NO_x	 O_3, NO_x, HC	 O_3, MET	 O_3, MET, NO_x, HC	 O_3, NO_x, HC, MET
Oregon	7					
Pennsylvania	14	5			3	14
Rhode Island	1			1		
South Carolina	7					
South Dakota						
Tennessee	8					
Texas	5			7	16	
Utah	4	4				
Vermont	3					
Virginia	11	7			1	
Washington	8					
West Virginia		2				
Wisconsin	20	4	3			2
Wyoming						
TOTAL	419	109	40	38	47	25

[a] Sites were counted as reporting NO_x if NO, NO_2, NO_x, or any combination was reported; HC if THCs, NMHC, CH_4, or any combination was reported; and MET if any meteorological parameter from Table 2 was reported.

combinations of pollutants. The general combinations of pollutants were used to reduce the number of possible combinations.

A review of Tables 3 and 4 indicates that approximately the same number of sites reported O_3 in 1980 (672) and 1981 (678). In 1980, the number of sites reporting data for meteorological parameters, HC, and NO_x was higher than the number for 1981. This is especially evident for Maryland, Massachusetts, New Jersey, New York, and Pennsylvania. During 1980, these and other states in EPA Regions I, II, and III participated in the Northeast Corridor Regional Modeling Project (NECRMP), a project involving the collection of data to assist states in developing control strategies for the 1982 Ozone State Implementations Plans.

Figure 1 present the locations of sites that reported O_3 data for 1981. As indicated by Table 3, these sites are concentrated around the most populous areas of the country, with only Idaho, South Dakota, and Wyoming reporting no data.

DATA AVAILABILITY

SAROAD is designed to store, analyze, and retrieve air quality and meteorological data. At least 30 different reports provide the user with quarterly or yealy summary statistics or individual hourly data values. The hourly data values are available in a hard copy format that displays data values for 1 mo per page or in a computer-readable format on magnetic tape. These

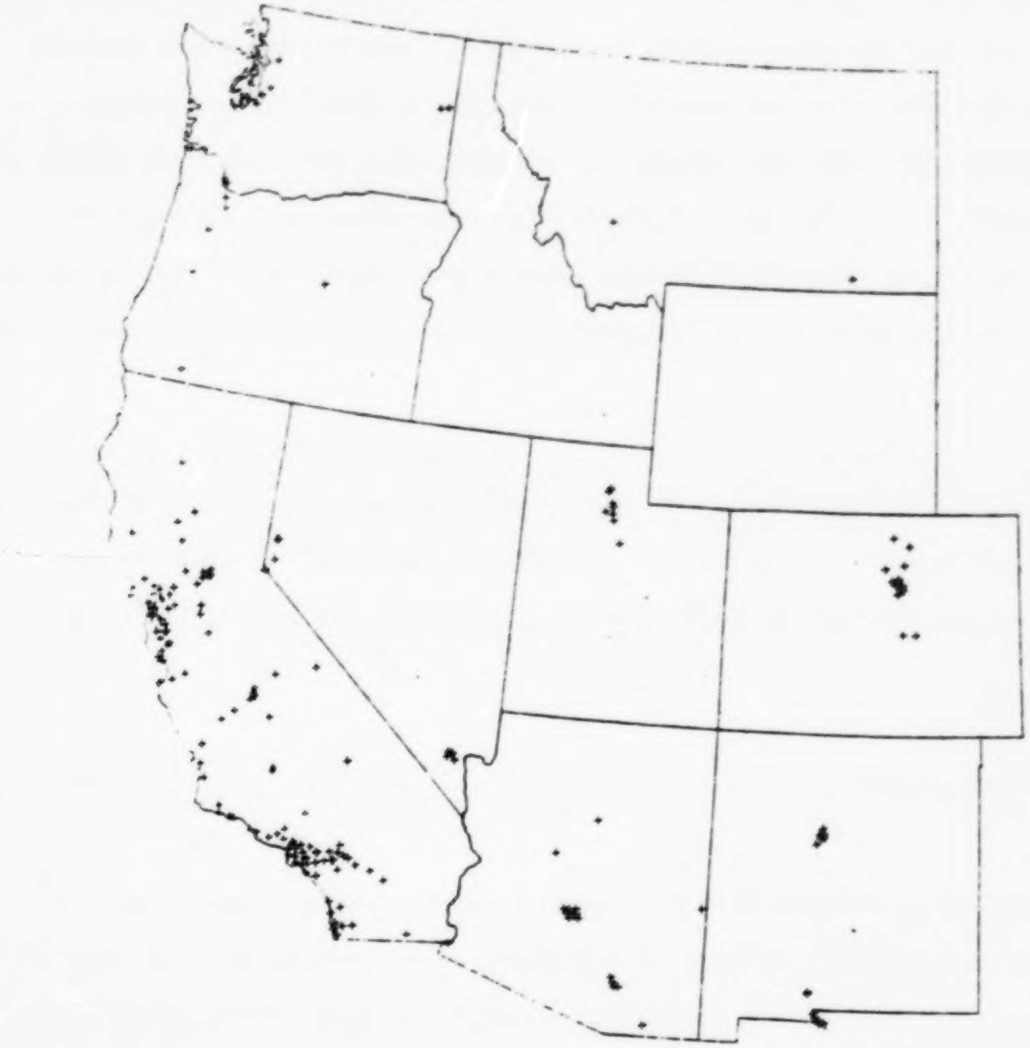

Figure 1. Sites reporting O_3 data for 1981.

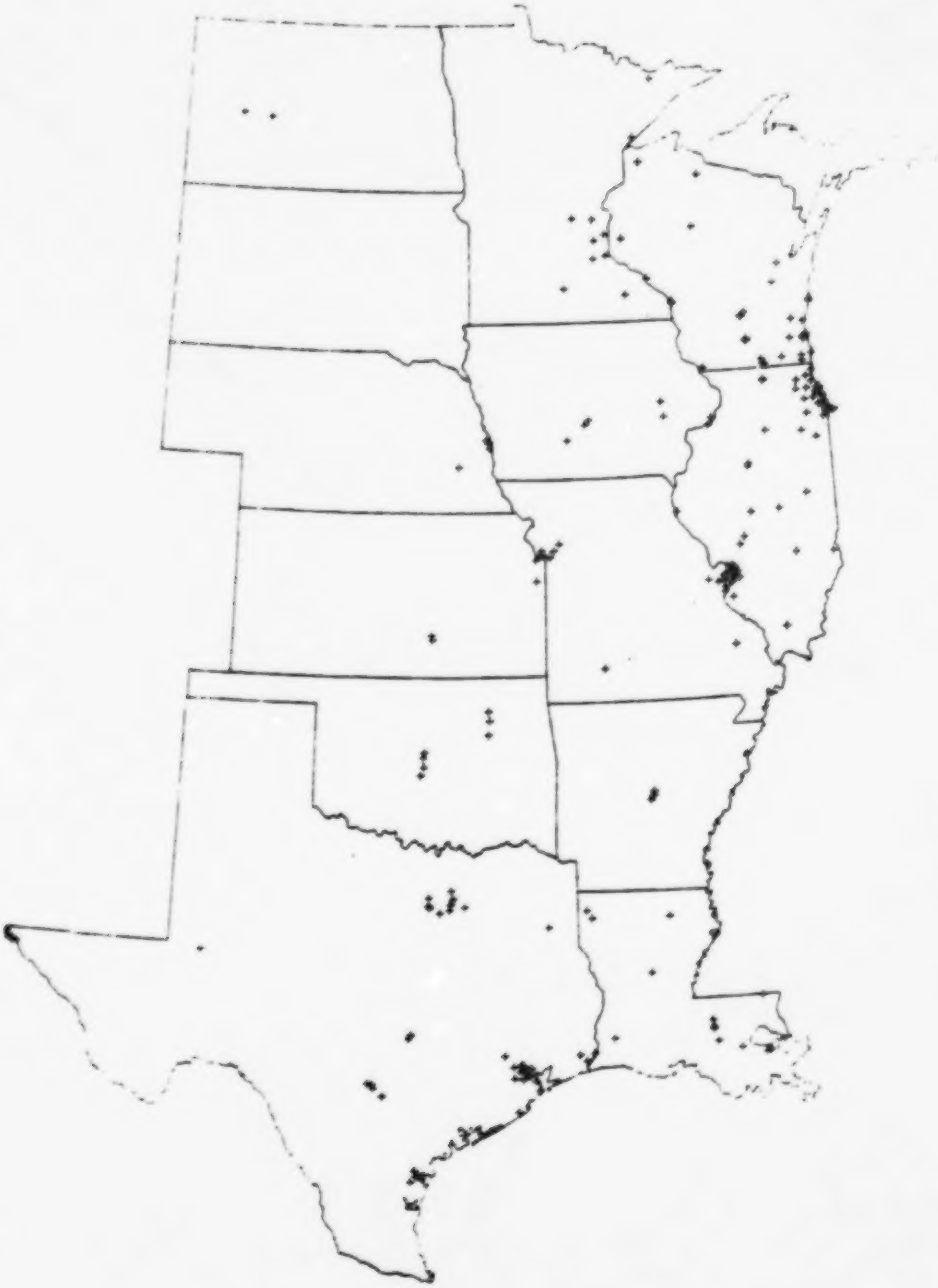

Figure 1. (continued).

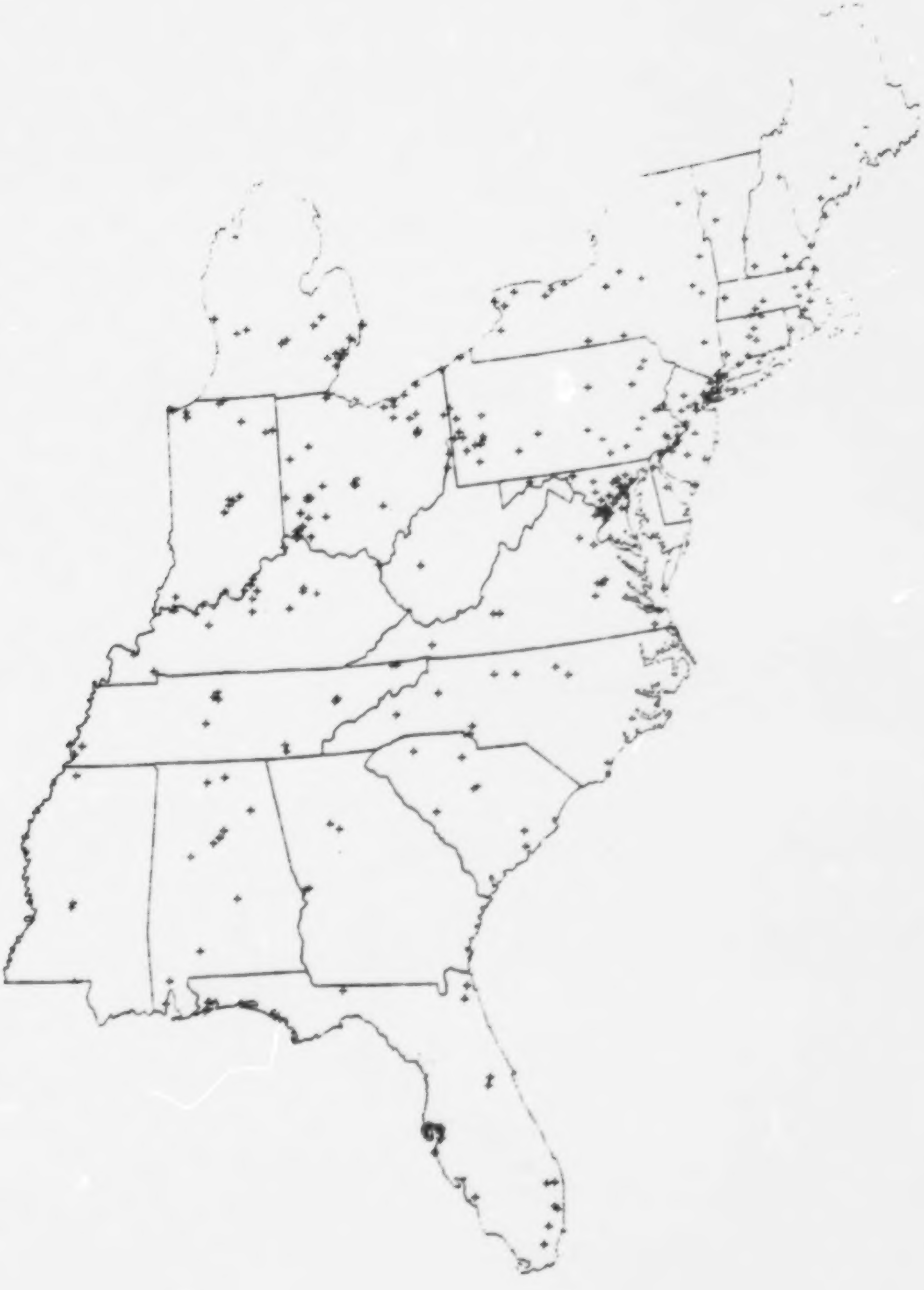

Figure 1. (continued).

reports permit data selection based on geographical area, pollutant, and time period.

Publications that identify the most useful report programs are available. Requests for publications and data may be addressed to the author.

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DISCUSSION

D. Jost: Does SAROAD allow for data extraction also? For example, if we are asking for the high and low concentrations during situations with high HC situations, do we need then to extract HC and O_3 data?

J. Summers: That could not be done directly. An overview would have to be done to identify high sites of O_3 or HCs and then retrieve the data separately. There are other studies that may have been done by other EPA groups that would already identify some of those, so that may already be available.

P. Misra: Do these reports give an estimate of the errors in these data?

J. Summers: As I mentioned, the precision-accuracy data were required to be reported beginning in 1981. When these reports were designed, of course, precision-accuracy reporting was required. So, we have a separate report after the report that gives the precision-accuracy data. Right now a working group is trying to evaluate the precision-accuracy data and come up with an exact way of how it can best be used.

So, although we have got about 2 yr of it already, we are not completely sure how it is going to be used.

APPENDIX. GUIDELINES FOR AEROMETRIC DATA PRESENTATIONS

Surface Air Quality

Data base name/source	Storage and Retrieval of Aerometric Data
Area of coverage	United States
Total number of monitoring sites	Approximately 700 ozone sites for 1981
Spatial distribution (attach site map)	
Year of record	1981
Check available site information:	
____ physical location (lat-long, UTM)	Both sets of coordinates
____ geographic location (state/province/ department, other sublevels)	State, county, city
____ elevation (MSL, AG)	Both
____ classification (i.e., urban, rural suburban, remote)	Station type combine this and dominating influence
____ environment of site	Only site address unless information in comments
____ descriptive information	
____ dominating influence (i.e., industrial residential, mobile)	See classification
____ other, specify:	
Parameters (attach table of measured parameters, associated equipment type/analysis method and temporal resolution)	

Upper Air Quality

Data base name/source	
Spatial distribution (attach standard of flight paths)	
Year/date of record	
Parameters (attach table of measured parameters, associated equipment type/analysis method and temporal resolution)	
Spatial resolution	

Surface Meteorology

Data base name/source	Storage and Retrieval of Aerometric Data
Area of coverage	United States
Total number of stations	Varies by parameters and year
Spatial distribution (attach site map)	
Site information available	Same as air quality
Parameters measured	Wind speed, direction, temperature, radiation
Time interval of measurements	Hourly averages
Year/date of record	1981

Data Quality

Is data suitable for model evaluation?	The air quality data is collected utilizing consistent sampling and analysis procedures and is suitable for model evaluation. Meteorological data is not reported by all States and is of unknown quality.
Are standard quality assurance procedures implemented?	Quality assurance procedures exist and were implemented in 1981 for all States and probably before 1981 for many States.
Comments on data reliability.	All procedures for sampling, data processing and analysis have been standardized and utilized for several years to ensure reliable data. Recent data are the most reliable.
List criteria for acceptance of data.	Sampling site must be registered and sampling performed utilizing EPA approved procedures. Study must be longer than three months and sites usually operate for several years.
Present summary of quantity of ozone measurements of available.	Quality of ozone data is good especially for 1981 - present as quality assurance continues to improve.

NORTHEAST CORRIDOR REGIONAL MODELING PROJECT: DATA BASE OF REGIONAL AMBIENT CHEMICAL AND METEOROLOGICAL MEASUREMENTS*

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INTRODUCTION

An extensive data base of regional ambient chemical and meteorological measurements is available for the Northeast United States from the Northeast Corridor Regional Modeling Project (NECRMP). The NECRMP is a multiphased program conducted by the U.S. Environmental Protection Agency (EPA) in conjunction with state/local air pollution control agencies to support the development and use of models to evaluate control strategies for reducing O_3 concentration levels in the Northeast. The geographical domain for NECRMP is shown in Figure 1. The focal point of this program is the combined application of the Regional Oxidant Model (ROM) (Lamb, 1983) and urban models for this region.

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*This paper has been reviewed by the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

†On assignment from the National Oceanic and Atmospheric Administration.

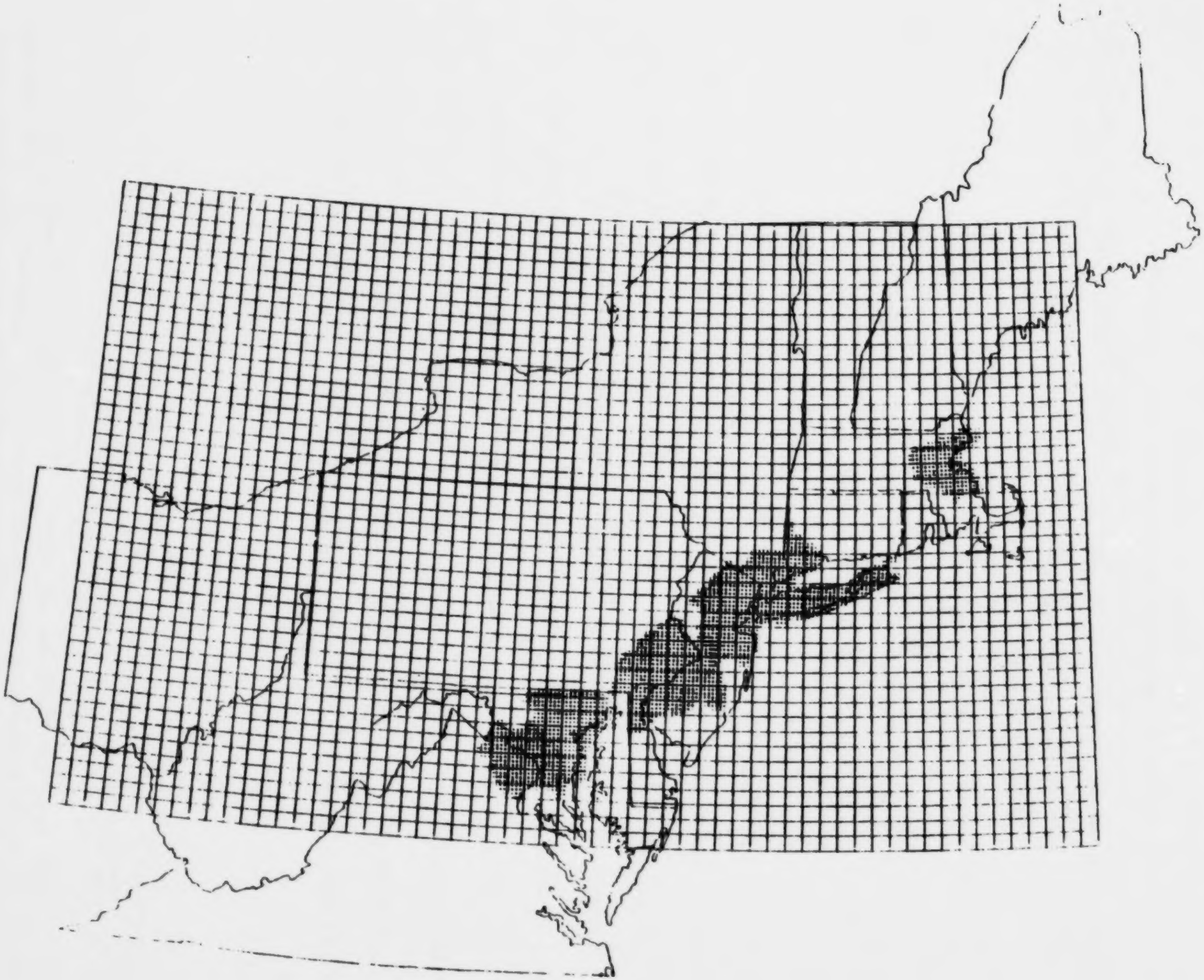

Figure 1. NECRMP regional/urban modeling domain.

The NECRMP ambient data base is the result of field measurement programs conducted in 1979 and 1980 to provide the chemical and meteorological measurements needed to develop and apply the ROM. These programs include the 1979 Northeast Regional Oxidant Study (NEROS I), the 1980 NEROS II, and the 1980 Urban Field Studies. Collectively, these studies have provided a variety of chemical and meteorological measurements on regional, urban, and site-specific scales. This paper discusses several of the regional-scale measurements and those urban-scale measurements that, when combined, also provide data on a regional scale. These include measurements aloft, obtained via instrumented aircraft; HC species measurements; meteorological measurements, obtained from the National Weather Service (NWS); and supplemental upper air meteorological measurements, obtained specifically for NECRMP. A companion paper at this conference by Summers (1983) discusses chemical and meteorological measurements available from surface networks operated by state/local agencies or contractors. Additional information on the scope of NECRMP and the ancillary field experiments is provided by Possiel et al. (1982).

REGIONAL DATA BASE COMPONENTS

Aircraft Measurements

Regional-scale chemical and meteorological measurements aloft were obtained during August 1979 as part of NEROS I. Instrumented aircraft operated by the Research Triangle Institute (RTI), Washington State University (WSU), and Brookhaven National Laboratory (BNL) provided continuous measurements of O_3, NO, NO_x, SO_2, light-scattering coefficient (b-scat), cloud condensation nuclei

(CCN), temperature, and relative humidity/dew point temperature. The specific instrumentation used for measuring these elements are listed in Table 1. During NEROS I, Regional Air Mass Characterization (RAMC) aerial sampling flights provided measurements at several altitudes within the daytime boundary layer and aloft above the nocturnal inversion at night. The typical sampling scenario, as

TABLE 1. NEROS I AIRCRAFT INSTRUMENTATION

Operator	Element	Equipment
Research Triangle Institute	O_3	Bendix 8002
	NO/NO_x	Monitor Labs 8440
	SO_2	Meloy 285
	CCN	Environment One Rich 100
	b-scat	MRI 1550 B
	AT[a]	Rosemount 102
	DPT[b]	EG & G 880
Brookhaven National Laboratory	O_3	AID 560
	NO/NO_x	TECO 14d
	SO_2	Meloy 165
	b-scat	MRI 1550
	AT	Yellow Springs 705
	DPT	EG & G 137-3C
	RH[c]	Weather Measure HM 111
	TSR[d]	Eppley pyranometer 8-48 A
	UVR[e]	Eppley UV radiometer
Washington State University	O_3	Bendix 8002
	NO/NO_x	Monitor Labs 8440 HP
	SO_2	TECO 43
	CCN	Environment One Rich 100
	b-scat	MRI 1550
	AT	Metrodata M-8
	RH	Metrodata M-8

[a]AT = air temperature.
[b]DPT = dew point temperature.
[c]RH = relative humidity.
[d]TSR = total solar radiation.
[e]UVR = ultraviolet radiation.

shown in Figure 2a, began with a traverse in the western portion of the domain at about 1200 EST, followed by four traverses at approximately 6-h intervals. These were performed at downwind distances specified to approximate Lagrangian sampling of the initial traverse. The traverses were approximately 500 km wide, flown in the wave pattern shown in Figure 2b. Occasionally, spirals were made from approximately 500 m to approximately 3,000 m to provide additional vertical resolution. In all, regional flights were conducted during six 24- to 48-h experimental periods.

Regional-scale chemical and meteorological measurements were also obtained via instrumented aircraft during 1980 as part of NEROS II and the Urban Field Studies. Instrumentation for the elements measured during these experiments are listed in Table 2. Aircraft were operated in Washington, DC, Baltimore, New York City, and Boston. Regional-scale sampling was conducted in a Lagrangian fashion similar to that for NEROS I. Urban-scale sampling was conducted in both Lagrangian and Eulerian modes in Columbus and Baltimore and in an Eulerian mode in the other Corridor cities. As shown in Figure 3, a Lagrangian sampling scenario typically began with measurements upwind of the city during the early morning, followed by a series of traverses perpendicular to the horizontal movement of a tetroon that had been released during the morning near the city. Traverses were performed at, below, and above the altitude of the tetroon at various distances downwind, to approximately 200 km from the city. In many cases, a forecast trajectory was used instead of a tetroon to estimate air parcel positions.

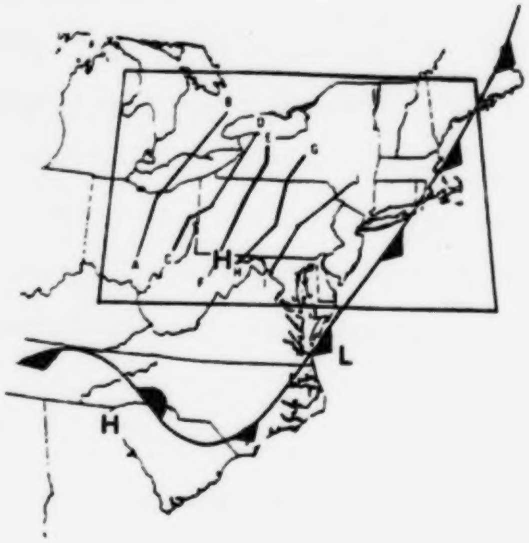

Figure 2a. Example of NEROS I regional sampling flight tracks.

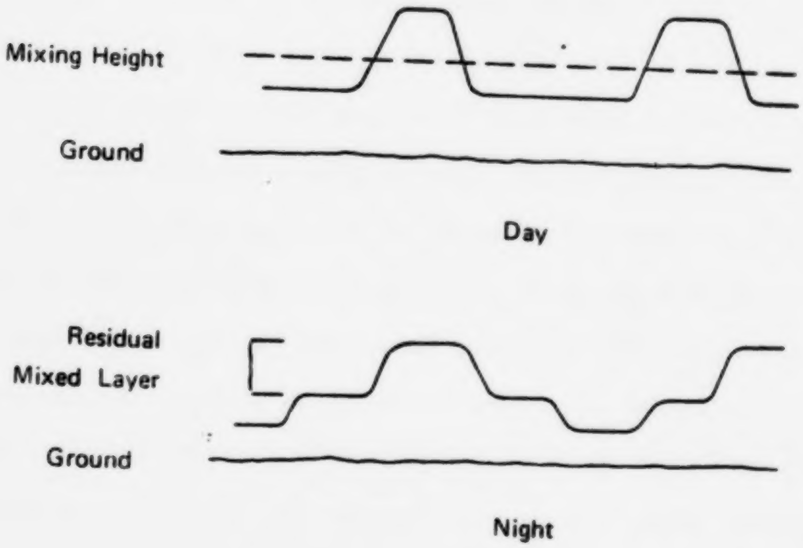
Mixing Height
Ground
Day
Residual
Mixed Layer
Ground
Night

Figure 2b. NEROS I regional flight patterns.

TABLE 2. AIRCRAFT INSTRUMENTATION USED DURING 1980 FIELD PROGRAMS

Operator	Element	Instrumentation
NEROS II Aircraft		
Environmental Monitoring, Inc.	O_3	Dasibi
	NO/NO$_x$	Monitor Labs 8440
	SO_2	Meloy 285
	SO_4	Meloy SA285
	b-scat	NA[a]
	AT[b]	NA
	DPT[c]	NA
AeroVironment	O_3	Bendix 8002
	NO/NO$_x$	Monitor Labs 8440
	SO_2	TECO 43
	SO_4	Not measured
	b-scat	NA
	AT	NA
	DPT	NA
Stanford Research Institute	O_3	Dasibi 1003 AAS
	NO/NO$_x$	Not measured
	SO_2	Meloy SA285E
	SO_4	Not measured
	b-scat	NA
	AT	NA
	DPT	NA
Washington, DC, Aircraft		
EPA-Las Vegas	O_3	Bendix 8002
	NO/NO$_x$	Monitor Labs 8440
	SO_2	TECO 43
	SO_4	Not measured
	b-scat	MRI 1550 B
	AT	Rosemount 102U2U
	DPT	General Eastern 1011

(continued)

TABLE 2. (continued)

Operator	Element	Instrumentation
Baltimore Aircraft		
Brookhaven National Laboratory	O_3	AID 560
	NO/NO_x	TECO 14d
	SO_2	Meloy 165
	b-scat	MRI 1550
	AT	Yellow Springs 705
	DPT	EG & G 137-3C
	RH[d]	Weather Measure HM 111
	TSR[e]	Eppley pyranometer 8-48 A
	UVR[f]	Eppley UV radiometer
Washington State University	O_3	Bendix 8002
	NO/NO_x	Monitor Labs 8440 HP
	SO_2	TECO 43
	CCN	Environment One Rich 100
	b-scat	MRI 1550
	AT	Metrodata M-8
	RIT	Metrodata M-8
New York City Aircraft		
Battelle Northwest Laboratory	O_3	Bendix 8000
	NO/NO_x	Monitor Labs 8440 HP
	SO_2	Not measured
	SO_4	Not measured
	b-scat	MRI 1550
	AT	Rosemount 102U2U
	DPT	EG & G 137-C

(continued)

TABLE 2. (continued)

Operator	Element	Instrumentation
Boston Aircraft		
Battelle Columbus Laboratory	O_3	Bendix 8000
	NO/NO_x	Monitor Labs 8440 HP
	SO_2	Not measured
	SO_4	Not measured
	b-scat	MRI 1550
	AT	Rosemount 102U2U
	DPt	EG & G 137-C

[a] AT = air temperature.
[b] NA = not available.
[c] DPT = dew point temperature.
[d] RH = relative humidity.
[e] TSR= total solar radiation.
[f] UVR = ultraviolet radiation.

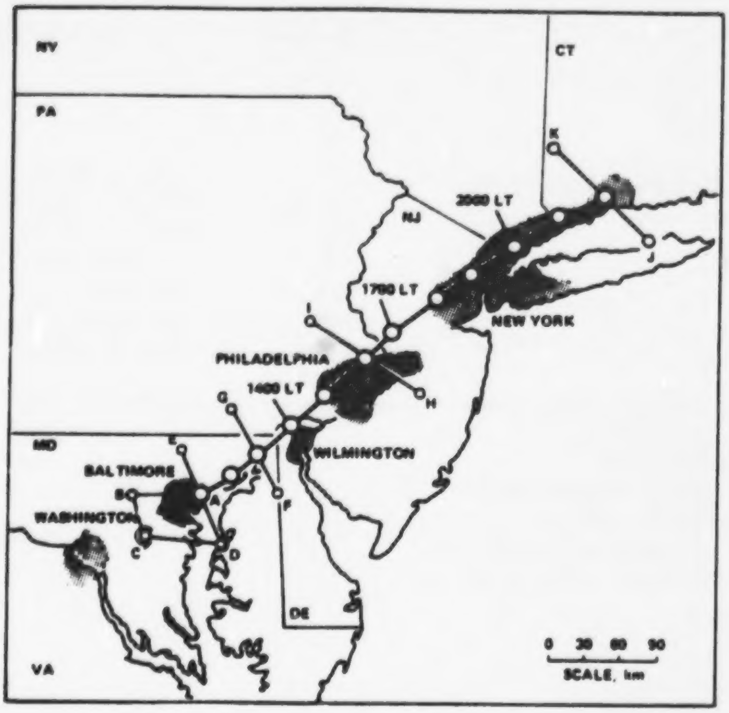
NY
CT
PA
NJ
K
2000 LT
J
1700 LT
I
NEW YORK
PHILADELPHIA
1400 LT
G
H
MD
E
WILMINGTON
BALTIMORE
B
A
F
WASHINGTON
C
D
DE
VA
0 30 60 90
SCALE, km

Figure 3. Typical Lagrangian sampling flight track. Times indicate position of tetroon; letters identify aircraft traverse locations.

The Eulerian flight scenarios included measurements 20 km to 40 km upwind between 0500 and 0600 EST, followed by a traverse and spiral pattern over the city and downwind from mid-morning through early evening (approximately 1700 EST). A typical afternoon Eulerian flight track is shown for New York City in Figure 4. Spatial resolution of the urban plume was obtained from traverses at several altitudes perpendicular to the plume, and/or traverses at a fixed, mid-boundary layer altitude perpendicular to the plume, with spirals to provide vertical resolution. On more flights, traverses were 50 km to 100 km wide. Also, Eulerian flights in Columbus were occasionally extended to include nighttime sampling of the urban plume. The total number of flight days for each city was 17 in Columbus, 13 in Washington, DC, 22 in Baltimore, 19 in New York City, and 15 in Boston. Of these, there were 2 days with flights in all areas, 5 days with flights in all Corridor cities, 12 days with flights in both Washington, DC, and Baltimore, and 8 days with flights in both New York City and Boston.

The quality assurance program for aircraft operations included audits of the chemical instrumentation onboard each aircraft. Audits were conducted in the field during the first days of the program. The audit results are given by Murdoch et al. (1979) for NEROS I and Arey et al. (1980) for the 1980 aircraft programs. Routine quality assurance procedures included zero and/or span checks of chemical instrumentation prior to, during, and/or following each flight. All gaseous chemical measurements are in parts per million by volume; other elements are in metric units.

Hydrocarbon Species Sampling

A major effort to sample for HC species was incorporated in NECRMP. Grab samples (1 to 3 min) were collected from aircraft during sampling flights, and 1-h integrated samples were collected at fixed surface sites. All samples were analyzed in the laboratory by gas chromatography (GC) for species concentrations. The species analyzed are listed in Table 3. Concentrations are reported as parts per billion of carbon.

TABLE 3. HYDROCARBON SPECIES ANALYZED BY GAS CHROMATOGRAPHY DURING NECRMP

000 ETHANE
001 ETHYLENE
002 ACETYLENE
003 PROPANE
004 PROPENE
005 I-BUTANE
006 N-BUTANE
007 1-BUTENE
008 I-BUTENE
009 T-2-BUTENE
010 C-2-BUTENE
011 I-PENTANE
012 N-PENTANE
013 2-METHYL-1-BUTENE
014 1,3-BUTADIENE
015 1-PENTENE
016 ISOPRENE
017 3-METHYL-1-BUTENE
018 2,2-DIMETHYL-BUTANE
019 CYCLOPENTENE
020 CYCLOPENTANE
021 4-METHYL-1-PENTENE
022 2,3-DIMETHYL-BUTANE
023 2-METHYL-PENTANE,
024 3-METHYL-PENTANE
025 1-HEXENE
026 N-HEXANE
027 T-2-HEXENE
028 C-2-HEXENE
029 METHYL-CYCLOPENTANE
030 2,2,3-TRIMETHYL-BUTANE
031 2,4-DIMETHYL-PENTANE
032 BENZENE
033 CYCLOHEXANE
034 2-METHYL-HEXANE
035 2,3-DIMETHYL-PENTANE
036 3-METHYL-HEXANE
037 1,3-T-DIMETHYL-CYCLOPENTANE
038 2,2,4-TRIMETHYL-PENTANE
039 1,3-C-DIMETHYL-CYCLOPENTANE
040 N-HEPTANE
041 METHYL-CYCLOHEXANE
042 ETHYL-CYCLOPENTANE
043 2,4-DIMETHYL-HEXANE
044 2,3,4-TRIMETHYL-HEXANE
045 UNKNOWN-1
046 UNKNOWN-2
047 UNKNOWN-3
048 UNKNOWN-4
049 UNKNOWN-5
050 UNKNOWN-6
051 UNKNOWN-7
052 UNKNOWN-8
053 UNKNOWN-9
054 UNKNOWN-10
055 TOLUENE
056 2,3-DIMETHYL-HEXANE
057 2-METHYL-HEPTANE
058 3-METHYL-HEPTANE
059 2,2,5-TRIMETHYL-HEXANE
060 DIMETHYL-CYCLOHEXANE
061 N-OCTANE
062 ETHYL-CYCLOHEXANE
063 UNKNOWN-11
064 UNKNOWN-12
065 UNKNOWN-13
066 UNKNOWN-14
067 UNKNOWN-15
068 UNKNOWN-16
069 UNKNOWN-17
070 UNKNOWN-18
071 UNKNOWN-19
072 UNKNOWN-20
073 ETHYL-BENZENE
074 P-XYLENE
075 M-XYLENE
076 STYRENE
077 O-XYLENE
078 N-NONANE
079 I-PROPYL-BENZENE
080 N-PROPYLBENZENE
081 M+P-ETHYL-TOLUENE
082 1,3,5-TRIMETHYL-BENZENE
083 O-ETHYL-TOLUENE
084 SEC-BUTYL-BENZENE
085 1,2,4-TRIMETHYL-BENZENE
086 1,2,3-TRIMETHYL-BENZENE
087 N-DECANE
088 METHYL-STYRENE
089 1,3-DIETHYL-BENZENE
090 1,4-DIETHYL-BENZENE
091 1,2-DIETHYL-BENZENE
092 UNDECANE
093 SUM OF UNIDENTIFIED COMPOUNDS
094 T-4-METHYL-2-PENTENE
095 C-4-METHYL-2-PENTENE
096 2-METHYL-1-PENTENE
097 2-METHYL-2-PENTENE
098 T-3-METHYL-2-PENTENE
099 C-3-METHYL-2-PENTENE
100 1,1,1-TRICHLOROETHANE
101 1-METHYL-CYCLOPENTENE
102 TRIMETHYL-CYCLOPENTANE
103 2,5-DIMETHYL-HEXANE
104 3-ETHYL-HEXANE
105 ALPHA-PINENE
106 BETA-PINENE
107 LIMONENE
108 DELTA-3-CARENE
109 BETA-PHELLANDRENE
110 ACETALDEHYDE
111 ACETONE
112 T-2-PENTENE
113 C-2-PENTENE

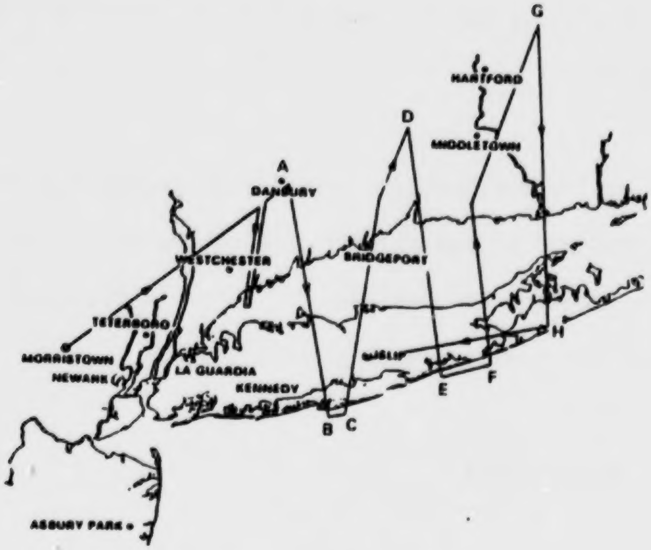
G
HARTFORD
D
MIDDLETOWN
A
DANBURY
WESTCHESTER
BRIDGEPORT
TETERBORO
MORRISTOWN
NEWARK
LA GUARDIA
KENNEDY
ISLIP
H
E
F
B
C
ASBURY PARK

Figure 4. Typical afternoon Eulerian flight track. Letters identify aircraft traverse locations.

were obtained within the mixed layer, the residual mixed layer at night, and above the mixed layer. Similar spatial sampling for HCs in the Northeast (but on a much smaller scale) was conducted during the 1980 NEROS II.

Hydrocarbon samples were also obtained over urban areas and within the urban plumes of Columbus, Washington, DC, Baltimore, New York City, and Boston during 1980. Aircraft grab samples were collected both within and above the surface stable layer on early morning flights and, in Columbus and Baltimore, within and above the mixed layer on later flights. Samples were collected upwind, over urban centers, and out to 100 km downwind. In all, 223 samples were analyzed for flights in Columbus, 45 in Washington, DC, 136 in Baltimore, 65 in New York City, and 87 in Boston. Also, as part of the 1980 program, over 162 quality assurance samples were collected for use in evaluating the comparability of ambient samples analyzed by the three GC laboratories operating during the program.

Ground-level 1-h samples were normally collected from 0500 to 0600 EST and from 0700 and 0800 EST at two sites in the urbanized portion of each city. In addition, upwind surface samples were collected in Columbus. A total of 794 surface samples were analyzed during the 1980 study.

Meteorological Measurements

NWS Surface Data--

Surface observations of meteorological conditions are made hourly at fixed locations by NWS. Figure 5 shows a plot of locations from which hourly data were received during the 1979 and 1980 field experiments. No distinction has been made between stations reporting on a 24-h basis and those reporting only during daylight hours. As indicated by the figure, NWS data were obtained not only for the NECRMP domain but also for the remainder of the contiguous United States and the adjacent areas of Canada and Mexico. The observations from these locations consists of a standard set of meteorological elements, some of which are determined with calibrated instruments and others that are subjectively determined by trained observers. The measured elements include temperature, dew point, station pressure, wind speed and direction, and the amount of precipitation. Elements that are subjectively determined are the sky cover, cloud type, and prevailing visibility. Some elements, such as the height of clouds, are either measured with instruments or are subjectively determined,

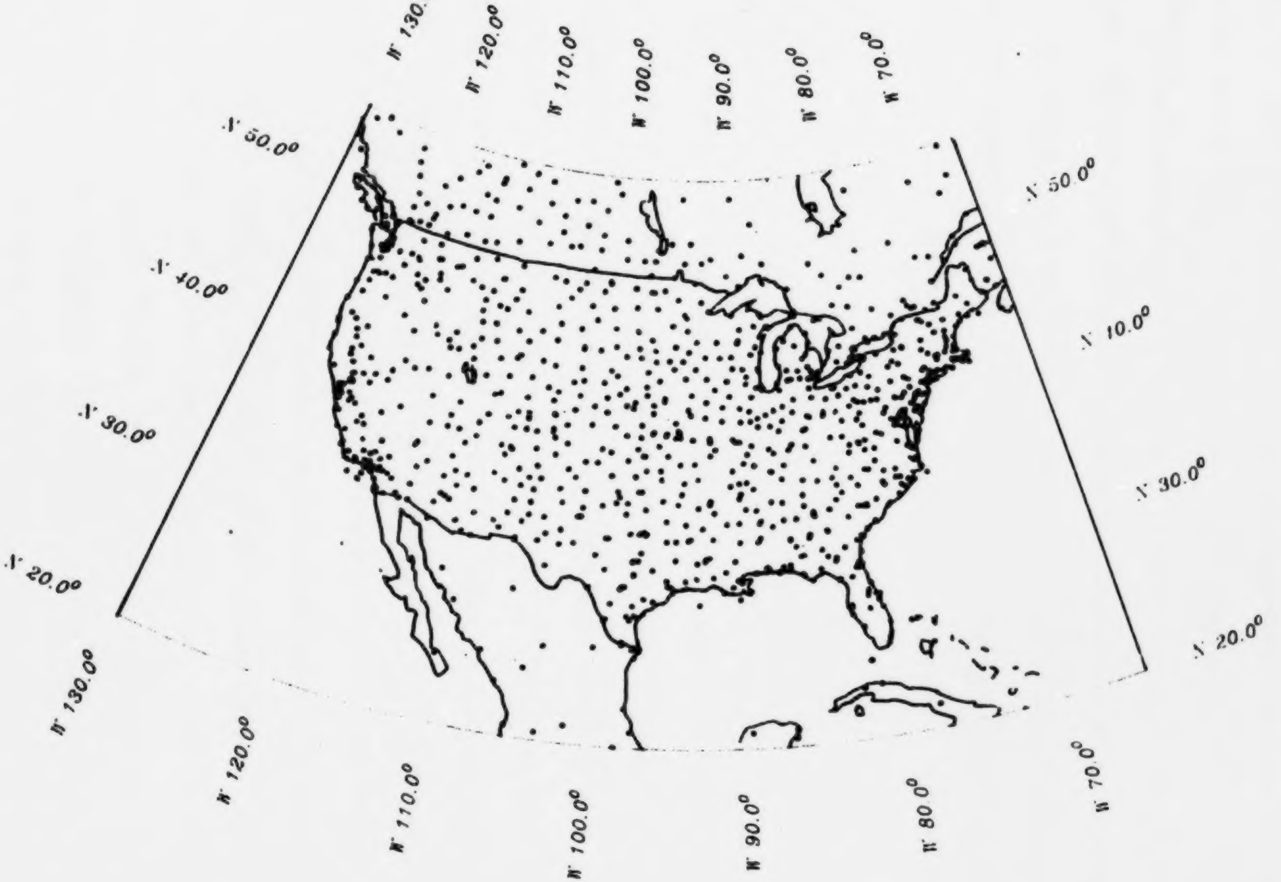
W 130.0°
W 120.0°
W 110.0°
W 100.0°
W 90.0°
W 80.0°
W 70.0°
N 50.0°
N 40.0°
N 30.0°
N 20.0°

Figure 5. Locations of weather stations transmitting hourly surface (airways) observations.

depending upon the value. Specific information on individual elements important to regional modeling is presented in Table 4 in the order in which these elements appear in the North American hourly airways sequence for data transmittal. All of the instruments used for meteorological measurements are maintained to operational calibration tolerances by a routine preventive maintenance program. The procedures for making, recording, and transmitting surface observations are described in detail in the Federal Meteorological Handbook, Number 1 Surface Observations (DOC), which is the standard reference for the United States. Other nations have similar documents.

NWS Upper Air Data --

In situ air data were taken by balloon-borne instruments packages twice daily at 0000 GMT (1700 EST) and 1200 GMT (0700 EST) on a routine basis by the rawinsonde stations shown in Figure 6. Additional soundings were conducted at 0600 GMT (0100 EST) and 1800 GMT (1300 EST) from July 15 through August 15, 1979, and from July 1 through August 31, 1980, at the stations within the NECRMP domain, except New York City. The following elements were reported at standard pressure surfaces: geopotential height (meters), temperature, dew point depression (degrees Celsius), wind direction (nearest five degrees of arc) and wind speed (to the nearest knot). Significant levels of temperature, dew point depression, and wind speed and direction are also reported, when required, to define the shape of the temperature/dew point profile. Details of operational procedures and quality assurance checks for the radiosonde and upper wind data are available in the Federal Meteorological Handbook, Number 3 (DOC).

TABLE 4. SURFACE METEOROLOGICAL DATA REPORTED BY NWS

Element	Comments
Cloud height	Measured by a ceilometer for altitudes up to approximately 5,000 ft; estimated for higher altitudes; units in feet above ground level.
Sky cover	Estimated using a rule of summation, which states that clouds at a higher altitude may not be reported as covering a smaller fraction of the sky than those at a lower altitude; reporting basis is tenths or fractions thereof.
Prevailing visibility	The greatest visibility that is equaled or exceeded throughout at least half of the horizon circle surrounding the observer; units in miles or fractions thereof.
Observed atmospheric phenomena	Hydrometeors (rain, snow, fog, drizzle, etc.) and lithometeors (haze, smoke, dust, etc.).
Sea level pressure	Air pressure measured by a barometer at the station analytically modified to be the pressure that would be observed if the station were at sea level; units in millibars.
Temperature/dew point temperature	Instantaneous measurements at time of observation from instrument in shelter or shielded housing; units in degrees Fahrenheit.
Wind direction/wind speed	Reported to the nearest 10 degrees of arc; units in knots.
Precipitation	Amount of hundredths of an inch reported at 3-h intervals.

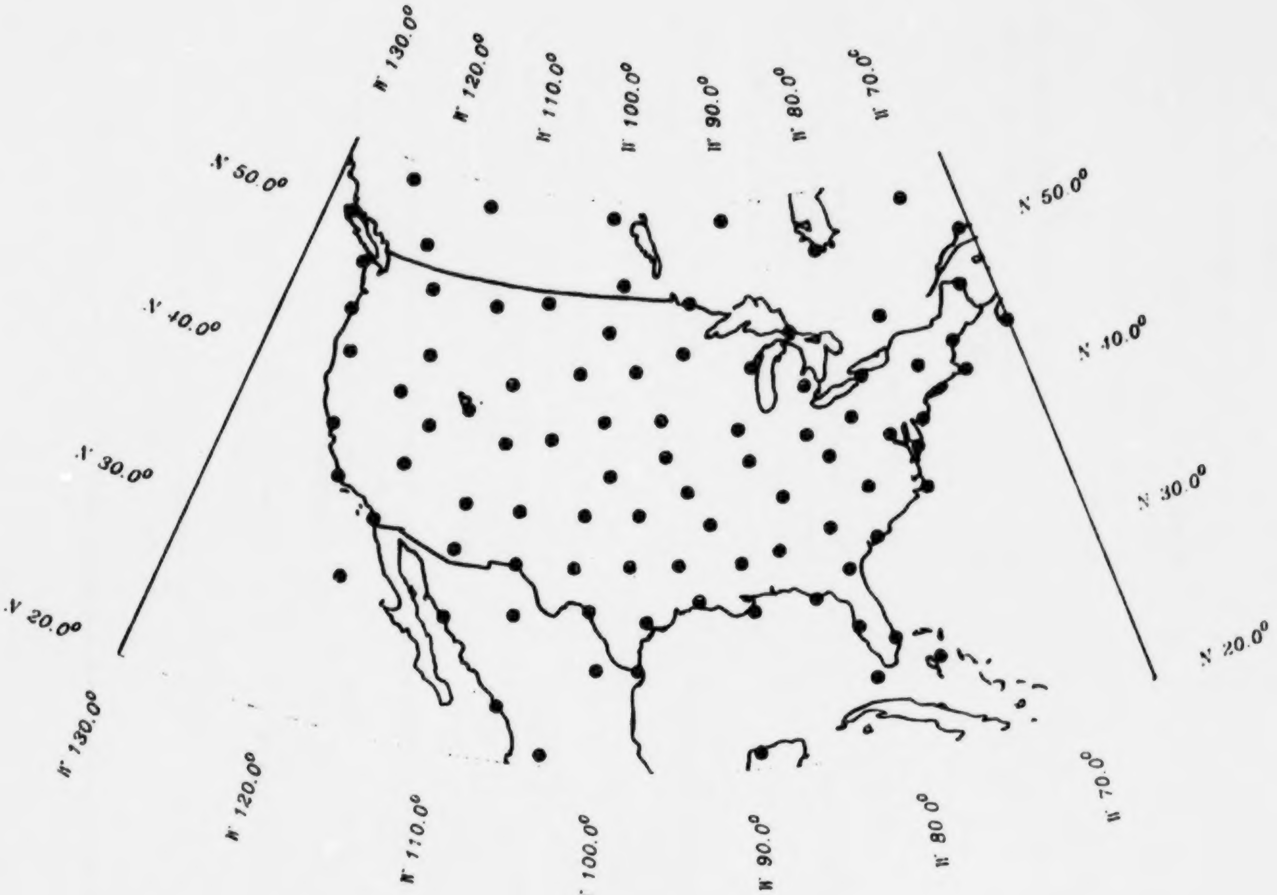
W 130.0°
W 120.0°
W 110.0°
W 100.0°
W 90.0°
W 80.0°
W 70.0°
N 50.0°
N 40.0°
N 30.0°
N 20.0°
N 50.0°
N 40.0°
N 30.0°
N 20.0°
W 130.0°
W 120.0°
W 110.0°
W 100.0°
W 90.0°
W 80.0°
W 70.0°

Figure 6. Locations of weather stations transmitting upper air (rawindsonde) observations.

NECRMP Supplemental Upper Air Data --

An intensive program of upper air meteorological measurements was conducted during 1980 to supplement existing NWS measurements. Basically, three types of measurement procedures were used: (1) rawinsonde soundings for temperature/dew point temperature and winds aloft; (2) pilot balloon observations for winds aloft; and (3) sodars for low-level mixing heights. The rawinsonde soundings were conducted at seven locations, sodars were operated at six locations, and pibal observations were taken at seven locations, as shown in Figure 7. Measurements were made at these sites from Juy 15 through September 12, 1980, except for all the pibals; the rawinsonde soundings were made at State College, Pennsylvania; and the sodars, which were terminated on August 15, were operated near Columbus.

The rawinsonde soundings at State College were conducted daily at 0100, 0700, 1300, and 1900 EST. Soundings at the other six locations were obtained 5 days per week, typically Monday through Friday, although the schedule was adjusted to obtain soundings on weekend days when aircraft flights were planned. The daily schedules of soundings at these sites are listed in Table 5. Slow-rise balloons were used for the soundings, and an average ascent rate of 400 ft/min was achieved on most soundings. Data were recorded between ground level and 700 mbar (approximately 3,500 m) only.

Temperature/dew point temperature and height values for mandatory and significant levels were determined by using standard NWS data reduction procedures, except that ±1/2°C rather than ±1°C was used for selecting

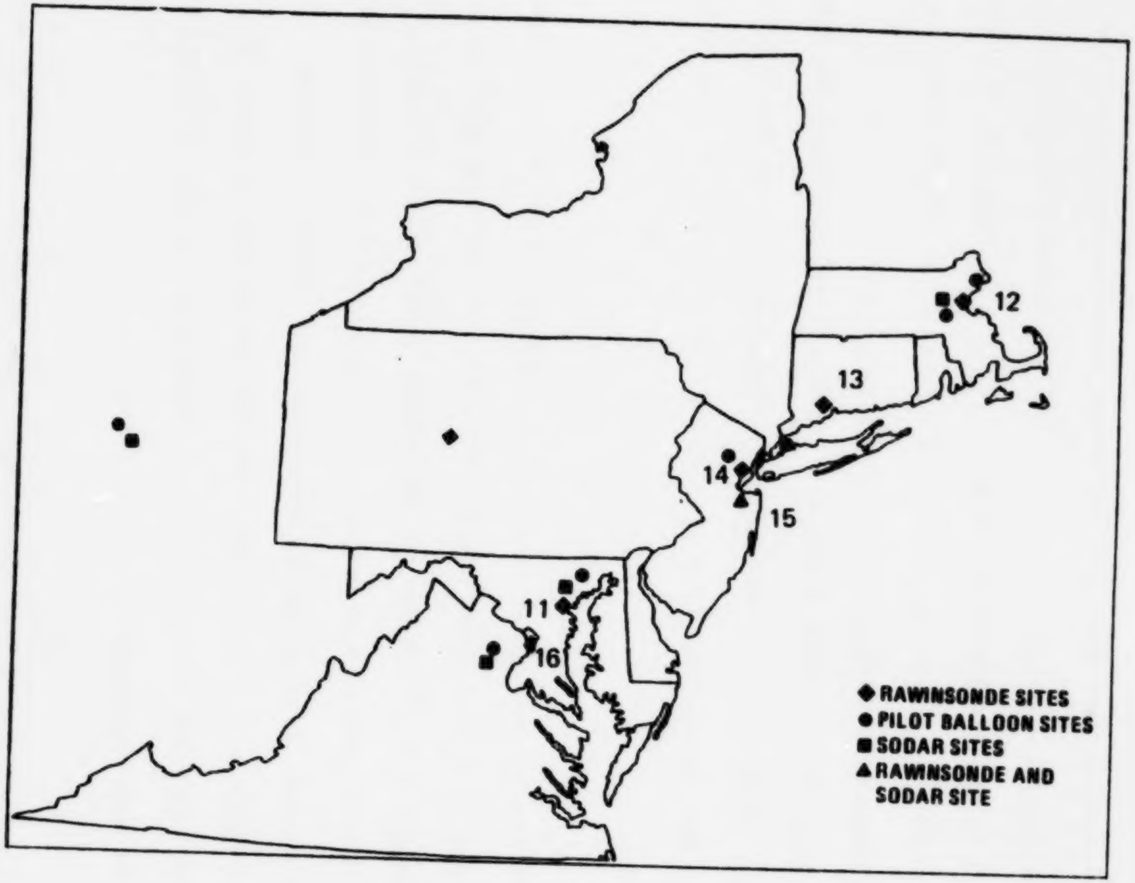
12
13
14
15
11
16
◆ RAWINSONDE SITES
● PILOT BALLOON SITES
■ SODAR SITES
▲ RAWINSONDE AND SODAR SITE

Figure 7. Locations of supplemental upper air meteorological measurement sites.

TABLE 5. SCHEDULE OF NECRMP SUPPLEMENTAL RAWINSONDE SOUNDINGS

Sounding Site	Start/End Date	Launch Time (EST)
Washington, DC (Site 16)	July 24-September 12	0500 0900 1300
Baltimore (Site 11)	July 16-September 12	0700 0900 1000[a] 1200[a] 1300 1400[a] 1500[a] 1600[a]
Marlboro (Site 15)	July 17-August 29	0500 0700 0900 1300
Newark (Site 14)	July 19-September 12	0500 0900 1100[b] 1300[c]
Derby (Site 13)	July 16-September 12	0700 0900 1100 1300 1500
Boston (Site 12)	July 18-September 12	0500 0700[d] 0900 1100[d] 1300 1500[d]

[a] Soundings on July 16 and 31 and August 1, 5, 6, 7, and 14 only.

[b] Soundings effective September 3.

[c] Sounding discontinued September 2.

[d] Winds only--no temperature measurements.

significant temperature points. The balloons were tracked by using LORAN-C navigational aids, and wind speed and wind direction were determined at 30-s intervals for the entire sounding. Standard NWS radiosondes were used for the temperature and humidity measurements.

Pilot balloon observations of wind speed and wind direction aloft were made almost daily during the period July 14 through August 15, 1980. Thirty-gram balloons were released hourly from 0400 EST through 1700 EST and tracked with a single theodolite. Readings of azimuth and elevation were recorded every 30 s for a total of 20 min. A constant-rise rate was assumed for reducing the raw data into wind speed and wind direction values. The reduced wind data were recorded at 110 m above ground level and continued at approximately 90-m intervals up to 3,690 m above ground level.

The AeroVironment model 300 monostatic acoustic radar (sodar) was used to provide a near continuous record of the thermal structure of the lower atmosphere from 30 m to 1,000 m AGL (the respective lower upper detectable limits of the instrument). Data from the sodar were reduced into 30-min average mixing height values determined as the height of the base of the lowest stable layer detected by the instrument.

Remotely Observed Data --

Satellite data were obtained during 1980 for the NECRMP domain from the National Oceanic and Atmospheric Administration. Cloud cover and cloud top heights have been produced by using the method of Reynolds and Vonder Haar (1977) for selected cases.

DATA BASE AVAILABILITY

The repository for the NECRMP data base will be the Environmental Sciences Research Laboratory, Meteorology Division, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina. At present, the various data base components, including those described in the paper, remain as separate entities. The data are currently undergoing various quality control checks and will be merged into a single chronologically ordered data base on magnetic tape. Additional information on the NECRMP data base can be obtained from Ms. Joan H. Novak, Mail Drop 80, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina 27711.

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DISCUSSION

P. Misra: In your aircraft measurements, do you have some kind of averaging procedure or are these instantaneous measurements?

N. Possiel: The aircraft measurements were made instantaneously and then averaged in 10-s to 25-s intervals, depending upon the individual aircraft.

P. Misra: It works out to some kind of an aerial average?

N. Possiel: It turns out to be some kind of an aerial average. The type of aircraft we used generally flew at about 130 knots, 120 to 130 knots, which is pretty standard.

S. Reynolds: You were out for about 1 mo in each of 2 yr. Were any interesting regional O_3 episodes observed while you were in the field?

N. Possiel: Yes. As a matter of fact, I showed you the regional sampling scenario from NEROS 1979, which had some very high O_3 concentrations transported across the region. During 1980, we were very fortunate in that O_3 was relatively high in that particular year, and we did obtain some very successful emissions in terms of seeing urban plume and regional transport.

A. Christie: You mentioned a meteorological data base. You have quite an extensive data base. When you are inputting that to the model, how is it going to be put in?

N. Possiel: I am not sure of the exact scheme. Perhaps Joan Novak can answer that.

J. Novak: As part of the modeling system, there is a series of preprocessors, which will take the data as it is received from the station. Then it will be transformed through the different meteorological preprocessors and actually put into the model in a series of matrix coefficients for submission and differential solving.

We do not just input the raw data as taken off the aircraft. We take each of the data sets independently and reformat it into the standard format for that type of data. Then we put it through extensive quality assurance/quality control procedures to validate the data, do extensive graphics to look at consistencies, or look for inconsistencies in a generated field. From that point we sort them, put them in chronological order, so they will be available to the model.

H. van Dop: In regard to this issue, I think that it is fruitful to keep in mind that it will be a difficult point to determine where the aerometric data ends and where the modeling starts. There is an interface between input data and a model, and that point of interface can be established more or less arbitrarily. Your preprocessing, does it belong to the model or does it belong to the input data? That is a difficult thing to distinguish.

CANADIAN SURFACE AIR QUALITY MONITORING NETWORKS*

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INTRODUCTION

The majority of continuous surface air quality measurements in Canada are carried out in urban-oriented air quality monitoring networks, of which the National Air Pollution Surveillance (NAPS) network is the largest. Some regional and background sites have been established to obtain daily measurements of SO_2, HNO_3, particulate nitrates, and trace elements by using filter pack techniques. Networks incorporating these measurements include CAPMON (Atmospheric Environment Service, Environment Canada) and APIOS (Ontario Ministry of the Environment). These will be discussed elsewhere. Regionally, representative monitoring sites for O_3 have been established only in southwestern Ontario and south-central Nova Scotia.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

NAPS Network

The NAPS network includes most of the air quality monitoring stations operating in Canada. Environment Canada coordinates the operation of the network and provides most of the monitoring equipment used. Actual day-to-day operation, calibration, and equipment maintenance are handled by municipal and provincial air pollution agencies.

The network was established in 1970 and was expanded until 1978, when it reached its present size. NAPS now consists of 500 instruments located in 52 cities.

Parameters Monitored and Site Locations--

Table 1 lists the stations, station addresses, and parameters measured in the NAPS network. (Geographical coverage is shown in Figure 1). Entries in the table indicate equipment ownership (F-Federal, P-Provincial) and sampling height (in meters) above the ground. The letter following the five-digit station code indicates land use at the sampling site (R-rural or residential, C-city core or commercial, I-industrial). A description of the monitoring equipment used, units of reported data, and sampling frequency are given in Table 2. Most of the sites listed have been in operation since 1976.

TABLE 1. LIST OF STATIONS IN NAPS NETWORK, 1981

		Parameters Measured[b]						
Station[a]	Location	SO_2	CO	NO_2	O_3	COH	Particulates and Pb	Dustfall and SO_3
	St. John's, Newfoundland							
10101C	Duckworth & Ordinance	F,09	F,09				F,09	
	Charlottetown, Prince Edward Island							
20101C	56 Fitzroy Street	F,10					F,09	
	Halifax, Nova Scotia							
30101C	Nova Scotia Technical College						F,12	
30102R	Dalhousie University						F,18	
30114R	Mt.St.Vincent University	F,10				F,10	F,10	
30115I	CFB Shearwater	F,08		F,08	F,08	F,08	F,11	
30116C	Barrington & Duke	F,02	F,02	F,02	F,02	F,02		
	Sydney, Nova Scotia							
30301R	Murphy Road							04
30309I	Pt. Edward, Richmond Plst							04
30310C	County Jail	F,04				F,04	F,02	04
30311R	Whitney Pier Fire Station	F,05				F,05	F,02	04
	Glace Bay, Nova Scotia							
30401R	Lake Road							04
30405C	General Hospital							04
30408R	South Street	F,03				F,03	F,02	
	Fredericton, New Brunswick							
40102C	York Street						F,07	
	Saint John, New Brunswick							
40201C	110 Charlotte Street						F,18	
40202C	Post Office	F,09	F,09	F,09	F,09			
	Moncton, New Brunswick							
40301C	774 Main Street						F,22	

(continued)

TABLE 1. (continued)

Station[a]	Location	Parameters Measured[b] SO_2	CO	NO_2	O_3	COH	Partic-ulates and Pb	Dust-fall and SO_3
	Montreal, Quebec							
50101R	Parc Jarry	F,03				F,03		05
50102R	Jardin Botanique	F,03		F,03	F,03		F,03	03
50103R	Pointe-Aux-Trembles	F,05	F,05		F,05	F,05	F,05	04
50104C	1125 Ontario Est	F,14	F,14	F,14	F,14	F,14	M,14	
50105C	1212 Drummond	M,13				F,13	F,11	
50106R	Ville St-Laurent	F,03				F,03	F,03	03
50107R	Ville Lasalle						F,04	04
50108R	1700 Bourassa, Longueuil						F,14	
50109C	Duncan & Decarie	P,05	F,05	F,05	F,05	F,05	P,03	
50110C	Parc Pilon, Mtl-Nord	P,05	P,05	F,05	F,05	F,05	P,05	
50111C	2900 Boul. Concorde	F,05	F,05	F,05	F,05		P,05	
50112C	Boul. Laurentides	F,03	F,03	F,03	F,03		P,03	
50113R	Pie X & Cardinal	F,05	F,05	F,05	F,05		P,05	
50114C	677 Ste-Catherine O		M,07					
50115C	Metcalfe & Maisonneuve	F,05	F,05	F,05	F,05	F,05	F,05	
50116R	3161 Joseph, Verdun	F,16	F,16	F,16	F,16	F,16	F,25	
	Hull, Quebec							
50203R	Gamelin & Joffre	F,05	F,05	F,05		F,05	F,05	
	Quebec, Quebec							
50301C	Parc-Autos Paq.Laliberte						F,15	
50302R	Parc Bardy						F,05	
50303I	Centre Loisirs Limoilou	F,15				P,15	F,12	
50304C	325 Dorchester Sud	F,05	F,05	F,05	F,05		F,12	
50306R	2026 Blvd. St-Cyrille	F,05			F,05		F,05	
	Sherbrooke, Quebec							
50402R	Casserne De Pompiers #5						F,09	
50403C	Wellington & Albert						F,17	
	Chicoutimi, Quebec							
50502R	Usine De Filtration						F,06	
50503C	222 Racine						F,06	
	Rouyn, Quebec							
50601C	Hotel de Ville	F,08					P,08	

(continued)

TABLE 1. (continued)

		Parameters Measured[b]						
Station[a]	Location	SO_2	CO	NO_2	O_3	COH	Particulates and Pb	Dustfall and SO_3
	Sept-Iles, Quebec							
50701C	Hotel de Ville	F,08					P,08	
	Trois-Rivieres, Quebec							
50801R	Hart & Ste-Cecile	F,05				P,04	P,06	
	Arvida, Quebec							
50901R	Powell & Hoopes	F,05					P,06	
	Tracy, Quebec							
51001R	225 Ave Courshesne	F,08						
51002R	Garneau & Rte 132						F,04	
	Thetford Mines, Quebec							
51101C	Boul, Ste-Marthe						F,05	
	Shawinigan, Quebec							
51201I	Frigon & Laval	F,05				F,05	F,05	
	Baie Comeau, Quebec							
51301R	39 Ave. Marguette	F,12					F,12	
	Ottawa, Ontario							
60101C	88 Slater Street	F,17	F,05	F,05	F,13	F,13	F,16	
60103C	Gilmour Street					F,12		
60104C	Rideau & Wurtemburg	P,04	F,04	P,04	P,04	P,04	P,04	
60105R	NRC, Montreal Road	F,05		F,05	F,05		F,05	
	Windsor, Ontario							
60201I	Morton Terminal Dock	P,04					F,04	
60202C	City Hall						P,15	
60203R	Tecumseh Water Works						P,03	
60204C	471 University Avenue	P,12	P,12	F,12	F,12	P,12	P,12	
60211R	College & Prince	P,04				P,04		
	Kingston, Ontario							
60301R	Queen's University						F,15	
60302R	Napier Street	F,06				F,06		

(continued)

TABLE 1. (continued)

		Parameters Measured[b]						
Station[a]	Location	SO_2	CO	NO_2	O_3	COH	Particulates and Pb	Dustfall and SO_3
	Toronto, Ontario							
60401C	67 College Street	P,16	F,04	P,19	F,19	P,19	P,17	19
60402R	Don Mills, Science Cntr.	P,09	P,09	F,09	P,09	P,09	P,09	
60403I	Evans & Arnold	P,04	F,04	P,04	P,04	P,04	P,01	01
60404R	5126 Yonge Street							05
60405I	John Street Pump Station							05
60406R	Rosehill Reservoir							04
60408C	Danforth & Robinson							05
60409R	Redlands Crescent	P,03				P,03	P,01	
60410R	Lawrence & Kennedy	P,05	F,05	P,05	P,05	P,05	P,04	
60412R	Bathurst & Wilson	P,06	F,06	F,06		P,06	P,01	
60413R	Elmcrest Road	P,04	F,04	P,04	F,04	P,04	P,04	
60414I	Sherbourne & Wilton St.	F,05	F,05	F,05	F,05	F,05	P,05	
60415R	Queensway W & Hurontario	F,04	F,04	F,04	F,04	F,04	F,04	
60416C	381 Yonge Street		P,04					
60417C	26 Breadalbane	P,15	F,15	P,15	F,15	P,15	P,13	
	Hamilton, Ontario							
60501C	Barton & Sanford	P,04	F,04	F,04	F,04	P,04	P,04	04
60502I	Burlington & Gage							05
60503I	Chatham & Frid						P,06	06
60505R	North Park	P,04		F,04		P,04	P,04	
60507C	Hughson & Hunter							10
60510I	Strathearn							05
	Sudbury, Ontario							
60602R	Ash Street	P,04	F,04	F,04	F,04	P,04	P,04	
60605C	19 Lisgar Street						P,11	
60606C	Kennedy Street	P,05						
	Sault Ste. Marie, Ontario							
60702R	Anna Mcrea Public School							05
60703I	Bayview & Young							06
60704C	Queen & Elgin	F,06						
60705R	550 Queen Street West						F,05	
	Thunder Bay, Ontario							
60801C	14 Algoma Street						P,12	12
60806R	435 James S.	F,17			F,17	F,17	F,17	

(continued)

TABLE 1. (continued)

Station[a]	Location	Parameters Measured[b]						
		SO_2	CO	NO_2	O_3	COH	Partic-ulates and Pb	Dust-fall and SO_3
	London, Ontario							
60901C	King & Rectory	P,04	F,04	P,04	P,04	P,04	P,04	04
60902C	372 Dundas					P,17	P,17	
	Sarnia, Ontario							
61004R	Front St. at C.N.Tracks	P,03	F,03	P,03	P,03	P,03	P,03	
	Petersborough, Ontario							
61103C	500 George Street	P,18				P,18	P,18	
	Cornwall, Ontario							
61201R	Memorial Park	P,04	F,04	F,05	P,04	P,05	P,04	
	St. Catharines, Ontario							
61301C	North St. & Geneva St.	F,06	F,05	F,06	F,06	F,06	F,06	
	Kitchener, Ontario							
61501C	Edna and Frederick	F,05	F,05	F,05	F,05	F,05	F,05	
	Oakville, Ontario							
61602R	Bronte and Woburn Cres.	P,05	P,05	P,05	P,05	P,05	P,05	
	Oshawa, Ontario							
61701R	Ritson Rd. & Olive Ave.	F,05	F,05	F,05	F,05	F,05	F,05	
	Guelph, Ontario							
61801C	Farquhar & Wyndham	F,04				F,04	F,04	
	Winnipeg, Manitoba							
70102R	Portage & Woodlawn						F,07	
70104I	Union Stock Yards						F,17	
70105R	Martin & Henderson Hwy							05
70110C	Kennedy & St. Mary's							04
70113I	Windermere & Rockman							03
70115C	Portage & Minto							06
70116I	Smith & King							03
70118R	Jefferson & Scotia	F,04	F,04	F,04	F,04	F,04	F,05	
70119C	65 Ellen Street	F,03	F,03	F,03	F,03	F,03	F,03	
70120R	604 St. Mary's Rd.	F,04	F,04			F,04	F,04	

(continued)

TABLE 1. (continued)

Station[a]	Location	Parameters Measured[b]						
		SO_2	CO	NO_2	O_3	COH	Particulates and Pb	Dustfall and SO_3
	Brandon, Manitoba							
70201C	11th St. & Princess Ave.						F,12	
	Regina, Saskatchewan							
80102R	3211 Albert Street						F,06	
80108C	12th Ave. & Smith St.						F,12	
80109C	1620 Albert Street	F,14	F,14	F,14	F,14	F,14	F,12	
	Saskatoon, Saskatchewan							
80203R	30th St. & 833 P Ave.						F,14	
80209C	Idylwyld Dr. & 33rd St.	F,11	F,11			F,11	F,11	
	Moose Jaw, Saskatchewan							
80301C	Fairford St. & 1st Ave.	F,15					F,12	
	Prince Albert, Saskatchewan							
80401C	1257-1st Ave. East	F,07					F,09	
	Edmonton, Alberta							
90121I	17 Street & 105 Avenue	F,05	P,04				F,04	
90122R	127 St. & 133 Avenue	F,04	F,04	F,04	F,04	F,04	F,04	
90125C	Prins Elizabeth & 108 St.							02
90126R	77 Avenue & 85 Street							04
90127I	115 Avenue & 159 Street							02
90128R	99 Avenue & 160 Street							03
90130C	10255 - 104th Street	F,09	F,09	F,09	F,09	P,09	F,06	
	Calgary, Alberta							
90204C	316-7th Avenue						F,09	
90218I	Bonny Brk & 18A St. S.E.	F,04				P,04	F,04	
90219C	620-7th Avenue, S.W.							09
90221R	Dalhousie Dr & Dalham Dr							02
90222R	39 St. & 29 Ave. N.W.	F,04	F,04	F,04	F,04	F,04	F,04	
90223C	11 St. & 38 Ave. S.E.							02
90224I	Ogdendale & 71 Ave. S.E.							02
90225R	Palliser Dr. & Oakwood S.W.							04
90226I	Sheppard & 84 Ave. S.E.							04
90227C	1611-4th Street, S.W.	F,06	F,06	F,06	F,06	P,06		

(continued)

TABLE 1. (continued)

Station[a]	Location	SO$_2$	CO	NO$_2$	O$_3$	COH	Particulates and Pb	Dustfall and SO$_3$
	Red Deer, Alberta							
90301C	4747 50th Street						F,08	
	Lethbridge, Alberta							
90501C	13 St. & 9 Avenue S.						F,15	
	Yellowknife, Northwest Territories							
99001C	50th Ave. & 49th Street						F,07	
	Vancouver, British Columbia							
00102R	100 Richmond Street					F,05	F,05	
00104R	27th & Ontario						F,18	
00106R	2294 West 10th Avenue	F,03	F,03	M,03	F,03	F,03	F,17	
00108I	250 West 70th Avenue		F,05	F,05	F,05	F,05	F,05	
00109C	970 Burrard						F,06	
00110R	E. Hastings & Kensington	F,05	F,05	F,05	F,05	F,05	F,04	
00111I	Rocky Pt. Park	F,04	F,04	F,04	F,04	F,04	F,04	
00112C	Robson/Hornby	F,05	F,05	F,05	F,05	F,05		
00113I	Annacis Island, Delta						F,04	
00114C	Municipal Hall, Richmond						F,14	
00115R	Newton Elem. Sch., Surrey						F,08	
00116R	Fire Hall, N. Vancouver						F,06	
00117R	Bcit Burnaby						F,12	
	Prince George, British Columbia							
00202C	1011 4th Avenue					F,18	F,18	
	Victoria, British Columbia							
00302C	1106 Cook St.	F,11	F,11	F,11	F,11	F,11	F,11	
	Kamloops, British Columbia							
00401C	301 Seyumour St.	F,15					F,11	

(continued)

TABLE 1. (continued)

		Parameters Measured[b]						
Station[a]	Location	SO_2	CO	NO_2	O_3	COH	Particulates and Pb	Dustfall and SO_3
	Whitehorse, Yukon							
09001C	Federal Building					F,04	F,08	

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[a] Letter following station code indicates land use, i.e., R = residential or rural, C = city core or commercial, I = industrial.

[b] Entries indicate equipment ownership (F = Federal, P = provincial) and sampling height above the ground (in meters).

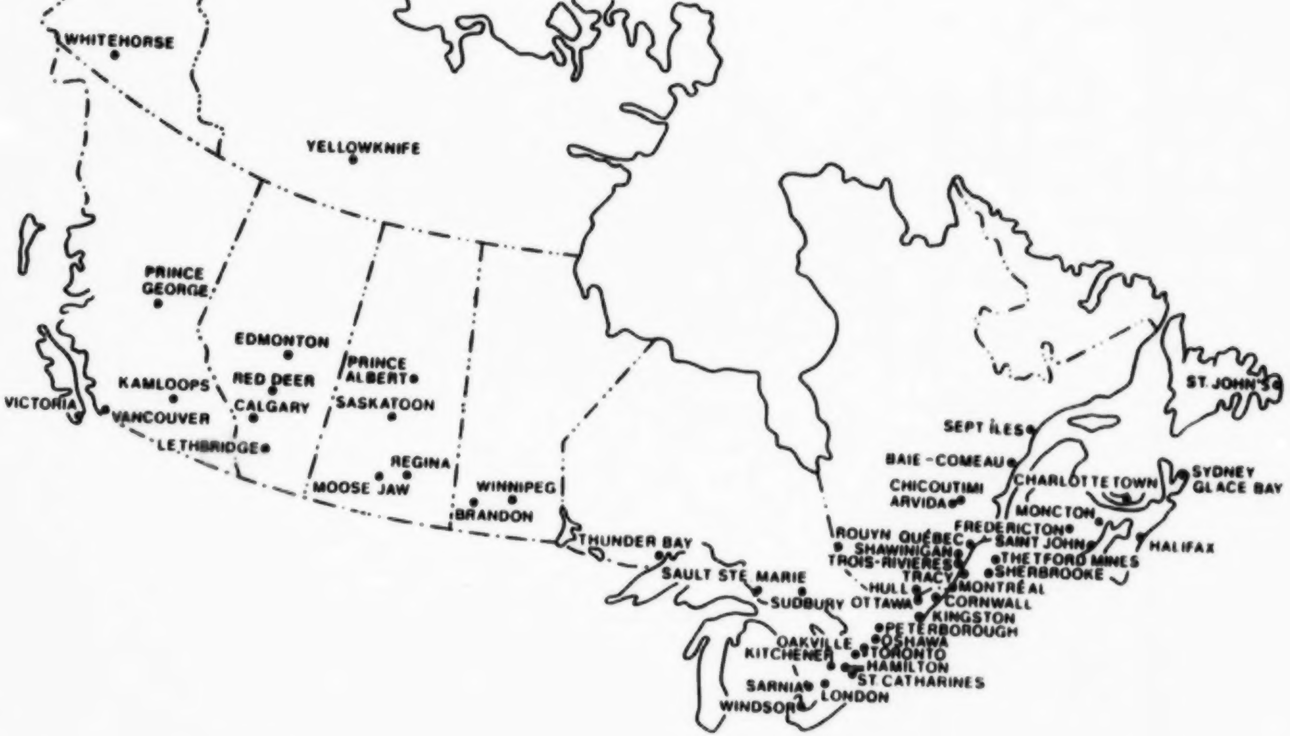
WHITEHORSE
YELLOWKNIFE
PRINCE GEORGE
EDMONTON
PRINCE ALBERT
RED DEER
KAMLOOPS
CALGARY
SASKATOON
VICTORIA
VANCOUVER
LETHBRIDGE
REGINA
MOOSE JAW
WINNIPEG
BRANDON
THUNDER BAY
SAULT STE MARIE
SUDBURY
OTTAWA
HULL
ROUYN
QUÉBEC
SHAWINIGAN
TROIS-RIVIERES
TRACY
MONTRÉAL
CORNWALL
KINGSTON
PETERBOROUGH
OSHAWA
TORONTO
OAKVILLE
KITCHENER
HAMILTON
ST CATHARINES
SARNIA
LONDON
WINDSOR
SEPT ÎLES
BAIE-COMEAU
CHICOUTIMI
ARVIDA
CHARLOTTETOWN
MONCTON
FREDERICTON
SAINT JOHN
THETFORD MINES
SHERBROOKE
HALIFAX
SYDNEY
GLACE BAY
ST. JOHN'S

Figure 1. National Air Pollution Surveillance Network (December 1981).

TABLE 2. DESCRIPTION OF NAPS NETWORK INSTRUMENTATION

Pollutant	Detection Principle	Unit of Measurement[a]	Concentration Reported	Type of Monitoring
SO_2	Coulometry Ultraviolet fluorescence	pphm	1 pphm	Continuous
CO	Nondispersive infrared spectrometry	ppm	0.5 ppm	Continuous
NO_2	Chemiluminescence	pphm	1 pphm	Continuous
O_3	Chemiluminescence Ultraviolet photometry	pphm	0.1 pphm	Continuous
Suspended Particulates (Soiling index)	Photometry	COH[a]	0.1 COH	Intermittent (12 2-h or or 24 1-h) samples daily)
TSP	Gravimetry	$\mu g/m^3$	1 $\mu g/m^3$	Intermittent (24-h sample every 6th day)
Pb (Particulate)	Atomic absorption spectroscopy X-ray fluorescence	$\mu g/m^3$	0.1 $\mu g/m^3$	Intermittent (24-h sample every 6th day)
Dustfall	Deposition by gravity	g/m^2/30 days	0.1/g/m^2/ 30 day	Intermittent (12 30-d samples per year)
Sulphation Rate	Reaction with lead dioxide	μg SO_3/100 cm^2/day	0.1 mg SO_3/ 100 cm^2/day	Intermittent (12 30-d samples per year)

[a] Coefficient of haze.

Site Descriptions--

Comprehensive site documentation is available for most sites in the network. Documentation includes geographic and street location, UTM coordinates, latitude and longitude, time zone, length of record, instrument details, scale of representativeness, land use by sector (within a 2-km radius of station), site elevation, airflow restrictions, manifold type, source influences on station (local, roadway, and major point sources), topographic map, and site photographs.

Data Quality--

A comprehensive quality assurance program has been established for the NAPS network. Monitoring and calibration equipment has been standardized, as have operational, zero/span, and multipoint calibration techniques. The Federal Government conducts an audit program for all continuous monitors reporting data to the network; data are rejected if the difference between monitor response and audit concentration is greater than ±15%. Compressed gas cylinders for zero/span calibrations are analyzed by a central Federal laboratory and distributed to all operating agencies. Validated hourly data submitted by the provincial and municipal cooperating agencies are subjected to a number of data-screening routines before incorporation in the data bank.

All archived data can be retrieved for any time period and distributed in magnetic tape format. Data are normally archived within 6 mo of collection.

ONTARIO MINISTRY OF THE ENVIRONMENT MONITORING NETWORK

The majority of monitoring instruments in the Ontario network have been incorporated into the NAPS network; however, supplementary monitoring data for NO, THC, and NMHC are available from the province. Additionally, a number of rural sites for O_3 have been established in the southwest portion of the province, as shown in Table 3 and Figure 2.

The province compiles site documentation information similar to that of the NAPS network. All monitoring is carried out under a very comprehensive quality assurance program incorporating all the elements of the NAPS program. Data are available in magnetic tape format for all sites, beginning with 1976 data.

ADDITIONAL OZONE MONITORING SITES

Supplementary O_3 monitoring data are available from a site in south-central Nova Scotia (Kejimkujic National Park), beginning with 1982 data, and for two sites within 30 km of Montreal (Tracy and Beauharnois).

TABLE 3. ONTARIO MINISTRY OF THE ENVIRONMENT--SUPPLEMENTARY MONITORING STATION

Region	Station Number	Station Address	UTM Grid		Air Intake Above Ground (m)	Pollutant Monitored			
			East	North		O_3	CO	HC	NO_x
Southwest	10001	College of Agriculture Tech., Huron Park W.	04600	47931	5	x			
	12008[a]	467 University Ave. W. Windsor	03316	46867	11	x	x	x	x
	13021	MOE Pump Station Middle Road Merlin	03991	46776	3	x			
	14064[a]	Centennial Park Front St/CN Tracks Sarnia	03854	47592	3	x	x	x	x
	14118	PUC Water Pump Station Highway 21 Petrolia	04027	47564	4	x			
	14903	Virgil LaSalle Froomfield Corrunna	03814	47520	3			x	
	14904	East Sombra P.S. Wilkesport	03892	47285	3	x		x	x
	15001[a]	King-Rectory London	04818	47595	4	x	x	x	x
	18007	Concession Road 2 Lot A Tiverton	04541	49053	4	x			
West Central	22071	Experimental Farm Simcoe	05597	47449	4	x			x
	22086	Cheapside Road 3 km S of Highway 3 Nanticoke	05821	47472	5				x

(continued)

TABLE 3. (continued)

Region	Station Number	Station Address	UTM Grid East	UTM Grid North	Air Intake Above Ground (m)	Pollutant Monitored O_3	Pollutant Monitored CO	Pollutant Monitored HC	Pollutant Monitored NO_x
	26029[a]	Edna/Frederick Street Kitchener	05427	48116	5	x	x		x
	27037[a]	North/Geneva Street St. Catharines	06431	47805	5	x	x	x	x
	29008[a]	North Park Hamilton	05984	47927	3			x	x
	29025[a]	Barton-Wentworth Hamilton	05939	47900	4	x	x		x
Central	31001[a]	67 College Street 5th Floor Toronto	06300	48352	20	x	x	x	x
	31086	15 Breadalbane Toronto	06302	48355	4	x	x	x	x
	31104[a]	26 Breadalbone Toronto API	06302	48356	15	x	x	x	x
	31105[a]	Sherbourne/Wilton Toronto	06317	48338	5	x	x		x
	31120	Junction Triangle Perth Avenue Toronto	06248	48344	9	x	x		x
	33003[a]	Lawrence-Kennedy Scarborough	06389	48452	3	x	x	x	x
	34002[a]	Science Centre Don Mills Road North York	06338	48419	9	x	x	x	x
	34007[a]	Bathurst-Wilson North York	06261	48437	2		x	x	x

(continued)

TABLE 3. (continued)

Region	Station Number	Station Address	UTM Grid East	UTM Grid North	Air Intake Above Ground (m)	Pollutant Monitored O_3	CO	HC	NO_x
	35003[a]	Elmcrest Road Etobicoke	06142	48338	4	x	x	x	x
	35033[a]	Evans-Arnold Etobicoke	06192	48302	3	x	x	x	x
	44008	Highway 2/North Shore Boulevard East Burlington	05972	47964	17	x			x
	44015[a]	Bronte Road/Woburn Cres. Oakville	06031	48059	5	x	x	x	x
	45025[a]	Ritson Road/Olive Avenue Oshawa	06724	48624	4	x	x		x
	46110[a]	Queensway West/ Hurontario Street Mississauga	06122	48249	5	x	x		x
	47035	M. of A. 509 Victoria Street, East Alliston	05918	48898	4	x			
	48002	MTC Yard, Highway 47 Stouffville	06391	48694	4	x			
	49010	Hwy 117/Paint Lake Road Dorset	06624	50096	3	x			
Southeast	51001[a]	McDonald Gardens Ottawa	04471	50312	4	x	x		x
	56051[a]	Memorial Park Cornwall	05208	49846	4	x	x		x

(continued)

TABLE 3. (continued)

Region	Station Number	Station Address	UTM Grid		Air Intake Above Ground (m)	Pollutant Monitored			
			East	North		O_3	CO	HC	NO_x
Northwest	63022[a]	Hospital 35 Algoma Street North Thunder Bay	03356	53672	22	x			
Northeast	71049[a]	Land Regulation Office Queen Street Sault-Saint Marie	07047	51543	6	x			
	71057	Michipicoten Avenue Lot 38 Town of Mission	06625	53110	4	x			
North	77016[a]	Ash Street Sudbury	04994	51486	3	x	x		x
NEMP	22901	100 M.S. Highway 59 Long Point Prov. Park	05502	47138	4	x	x		x
	22902	Tyneside/Chippewa Road Binbrook West	05914	47746	5	x	x		x
	22903	Cheapside Road/Walpole Conc. 5 Nanticoke	05821	47472	4			x	
	22904	Walpole South PS Sandusk Road Nanticoke	05794	47434	4			x	
	22905	Nanticoke Road/ Walpole Conc. 5 Nanticoke	05744	47456	4			x	

[a] Part of NAPS network.

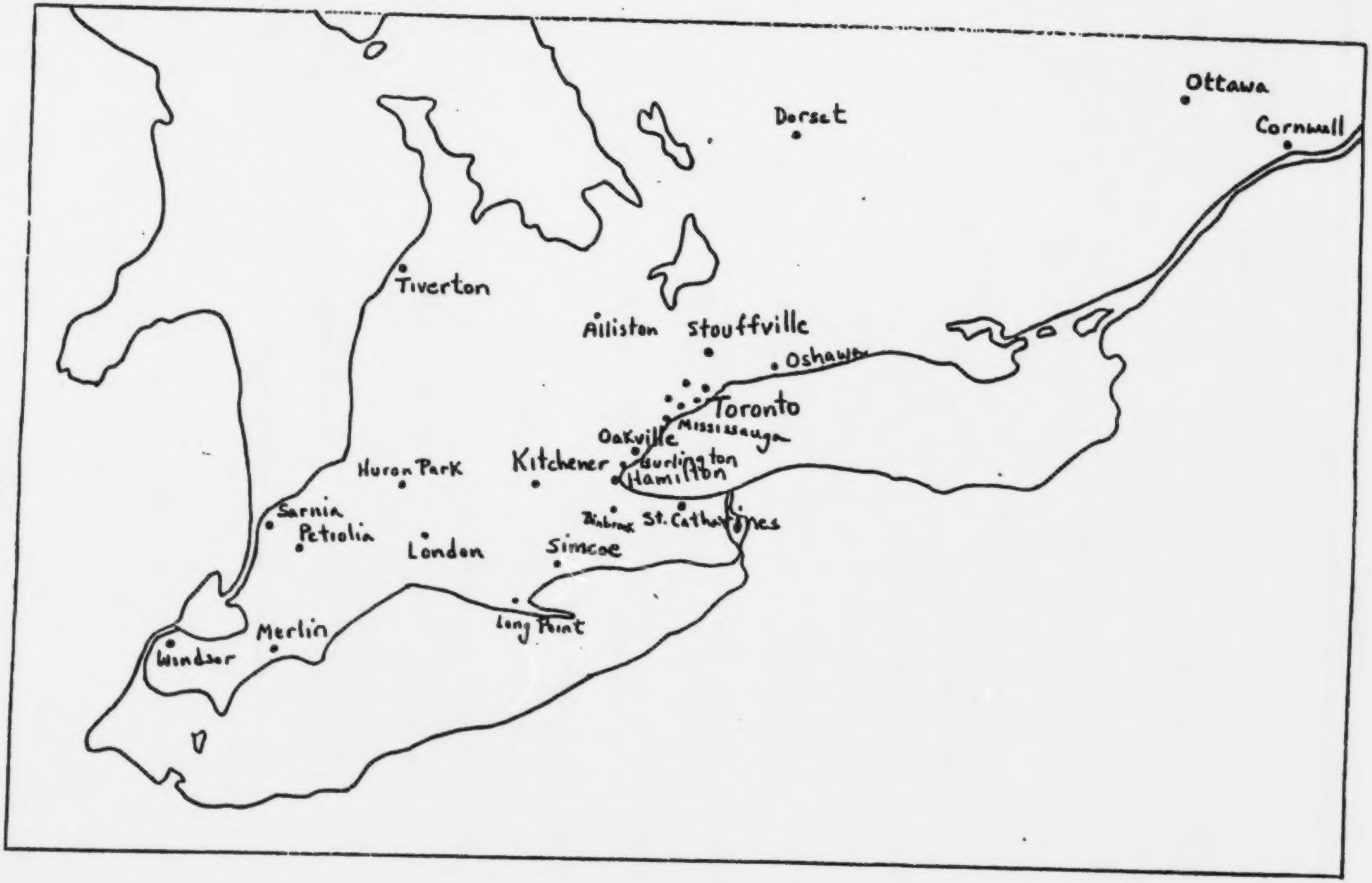
Ottawa
Cornwall
Dorset
Tiverton
Alliston
Stouffville
Oshawa
Toronto
Mississauga
Oakville
Burlington
Hamilton
Kitchener
Huron Park
Sarnia
Petrolia
London
Simcoe
Binbrook
St. Catharines
Long Point
Merlin
Windsor

Figure 2. Location of O_3 monitoring sites in Southern Ontario (1981).

AEROMETRIC DATA BASES IN THE NETHERLANDS*

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INTRODUCTION

Major air quality data bases in The Netherlands are available from the National Air Quality Monitoring Network and from the FLAT network. The former network, which is operated by The Netherlands Institute of Pulic Health, RIV (RIV, 1982; Van Egmond en Onderdelinden, 1981), comprises a number of fully automated, fixed monitoring stations that provide hourly averaged concentrations, 24 h a day, for NO and NO_2 (92 stations), O_3 (30 stations), CO (41 stations), and SO_2 (220 stations). The SO_2 and CO concentrations are measured by coulometry; the NO, NO_2, and O_3 concentrations are measured by chemiluminescence.

These measurements are made at a height of 3.8 m; at one station, measurements are made at 100 m and 200 m. Figures 1 and 2 show the stations for NO_x and O_3. The network for O_3, which has been in operation since 1978, comprises 30 stations that can be characterized as rural or semirural. For the other components, urban, suburban, industrial, and rural stations are available.

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*This report has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

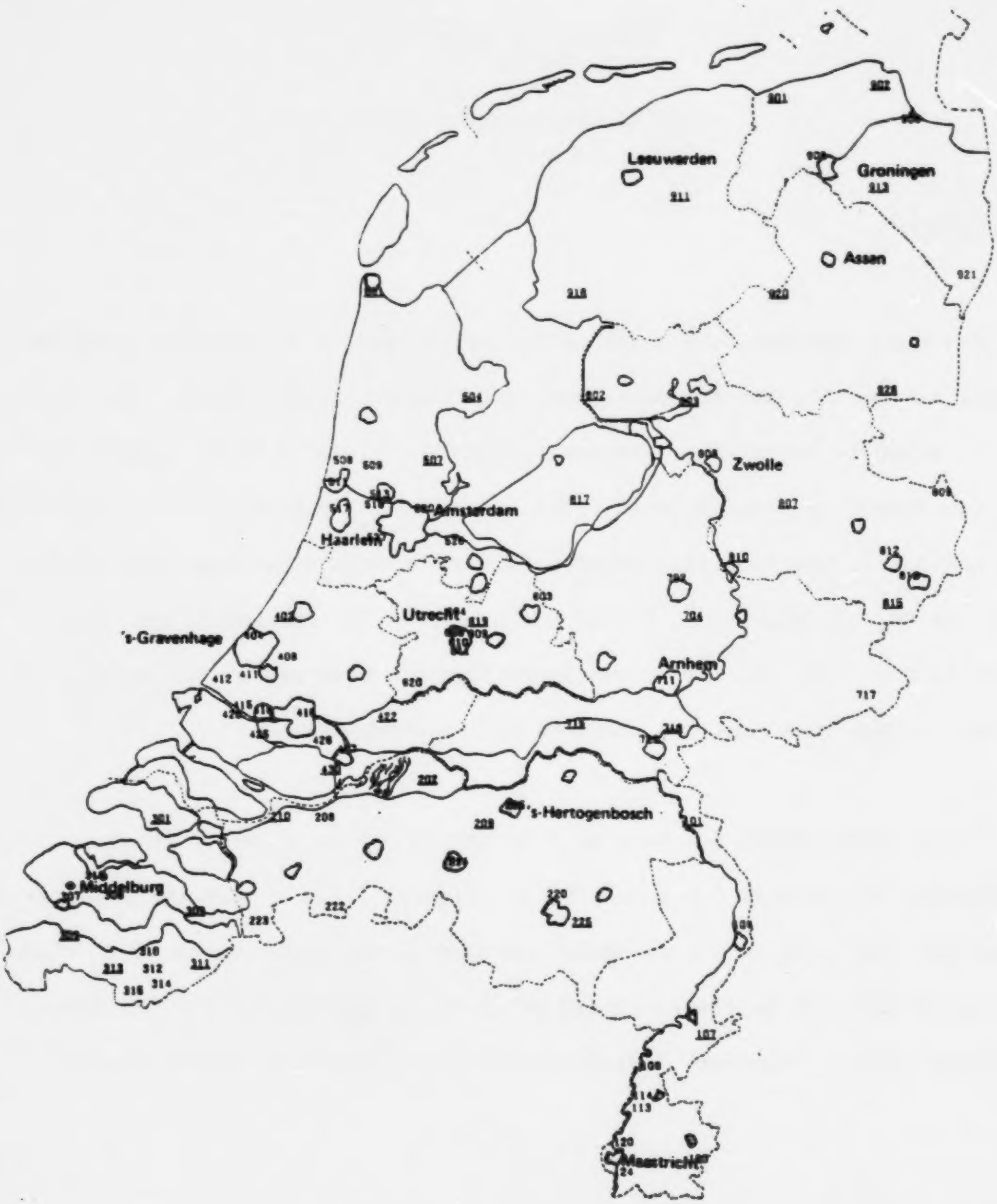

Leeuwarden
Groningen
Assen
Zwolle
Amsterdam
Haarlem
Utrecht
's-Gravenhage
Arnhem
's-Hertogenbosch
Middelburg
Maastricht

Figure 1. National Air Quality Monitoring Network: Measuring stations for NO_x.

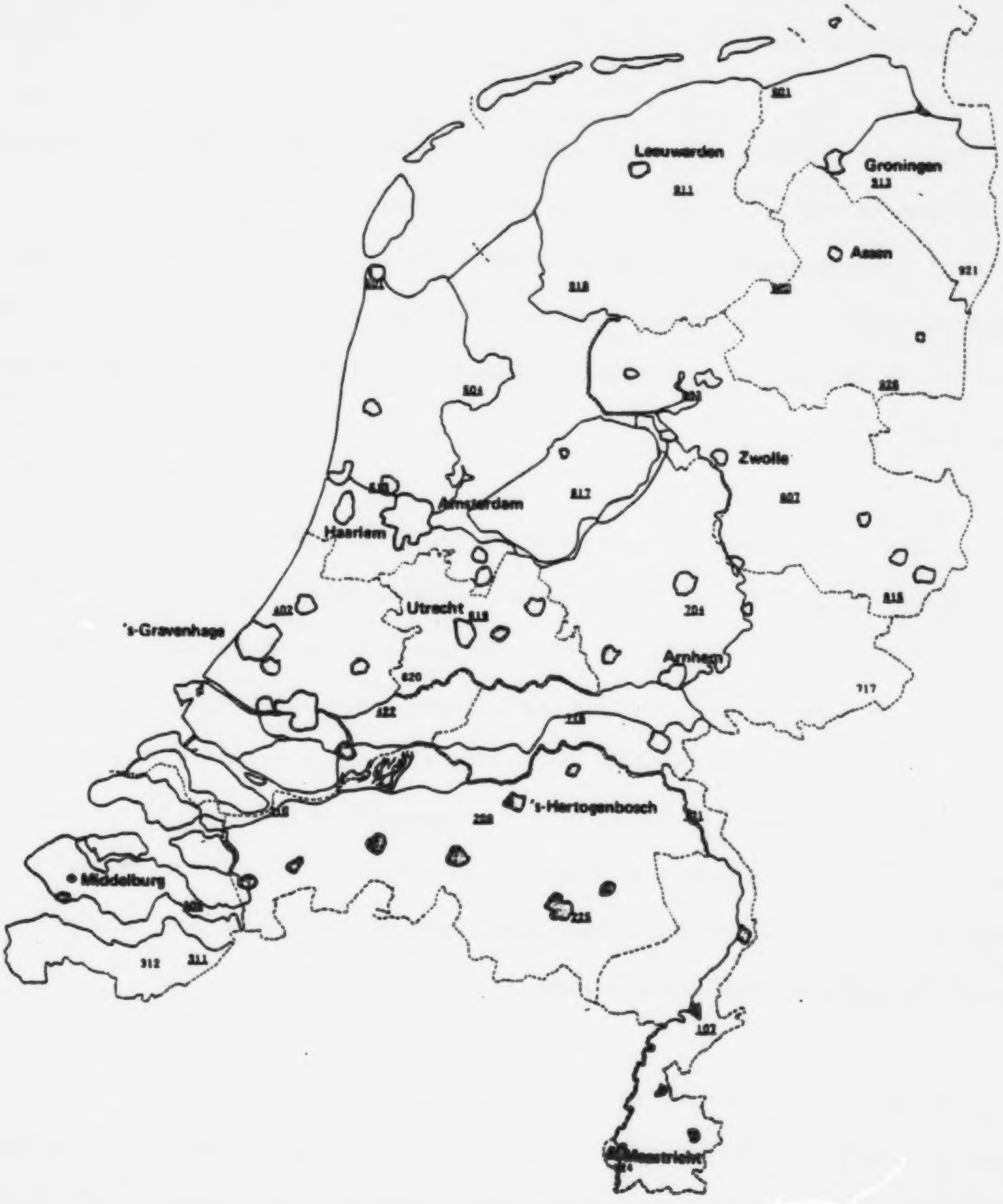

Leeuwarden
Groningen
Assen
921
Zwolle
Amsterdam
Haarlem
Utrecht
's-Gravenhage
402
Arnhem
620
717
's-Hertogenbosch
Middelburg
312
Maastricht

Figure 2. National Air Quality Monitoring Network: Measuring stations for O_3.

These fixed stations are supplemented with mobile vans that measure not only concentrations of the components mentioned above but also horizontal fluxes of NO_2 and SO_2 inflow by Barringer correlation spectrometry. Incidental airplane measurements are also carried out.

Future development of this network is aimed on the one hand at reducing its density and on the other hand at improving its flexibility with regard to the choice of component and sampling strategy. There is a need for data on HCs, sulphates, nitrates, and other components during air pollution episodes or under selected meteorological conditions. Remote operation of the sampling stations is under development.

A second major network, operated by TNO is called FLAT. This is the Dutch acronym for the project on Photochemical Air Pollution, Aerosols, and Toxicity, carried out by TNO from 1979 to 1982. The network was etablished for the specific purpose of evaluating mathematical air pollution models (Van Aalst and Guiherit, 1980). Photochemical precursors and products, aerosol components, and physical aerosol parameters were measured at six stations from 1979 to 1981. These are shown in Figure 3 and listed in Table 1 (Diederen et al., 1981). Table 2 summarizes the parameters measured, the temporal resolution, the sampling frequency, and the monitoring period.

In addition to these two major data bases, O_3 (half-hour) and PAN (quarter-hour) concentrations have been measured by TNO at Delft, the former since 1971 and the latter since 1973 (TNO, 1978; Heidema et al., 1981). All of these data are suitable for model evaluation. Standard quality assurance

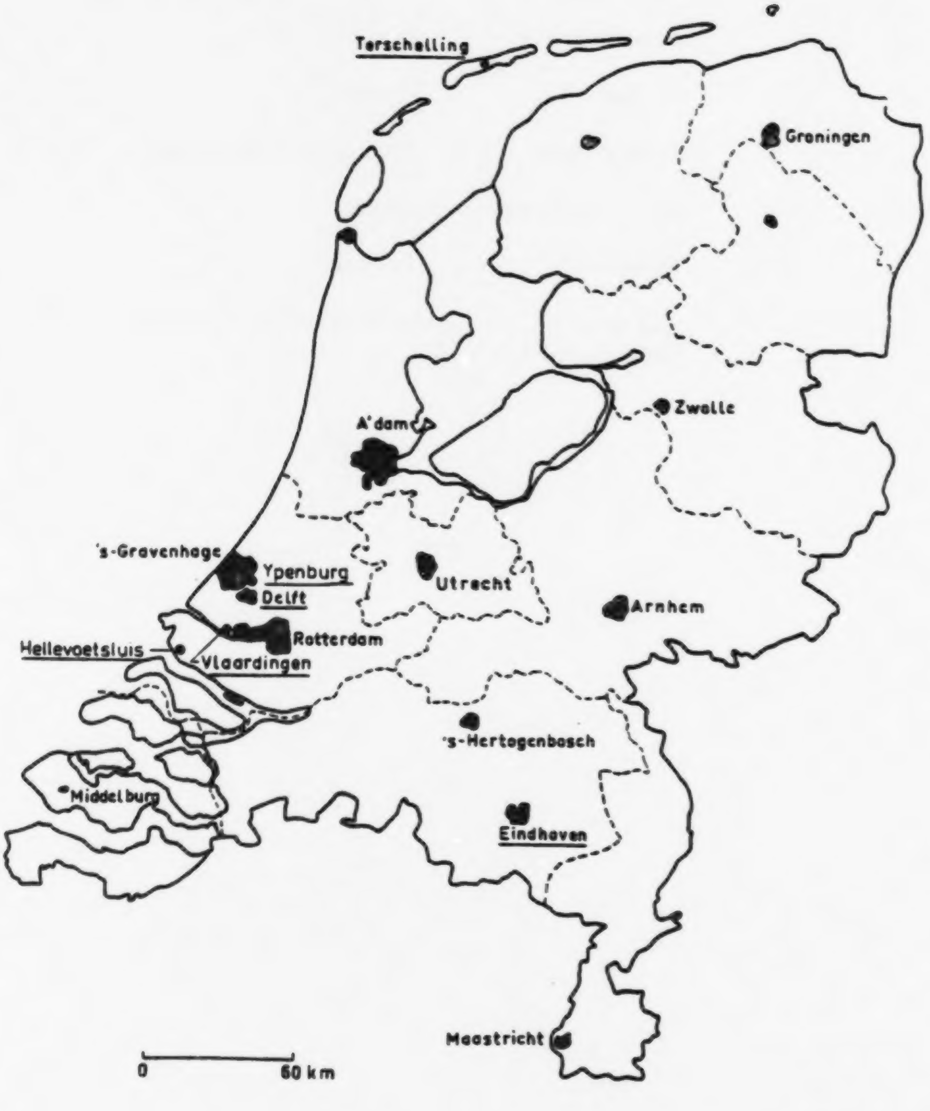

Terschelling
Groningen
Zwolle
A'dam
's-Gravenhage
Ypenburg
Delft
Utrecht
Arnhem
Rotterdam
Hellevoetsluis
Vlaardingen
's-Hertogenbosch
Middelburg
Eindhoven
Maastricht
0
60 km

Figure 3. FLAT monitoring stations (underlined) in The Netherlands (see Table 1).

TABLE 1. FLAT MONITORING STATIONS[a]

Station	Type
Delft	Suburan
Terschelling	Rural
Eindhoven	Urban
Vlaardingen	Suburban/industrial
Hellevoetsluis	Rural
Ypenburg	Suburban

[a] See also Figure 3.

TABLE 2. PARAMETERS MEASURED AT FLAT STATIONS

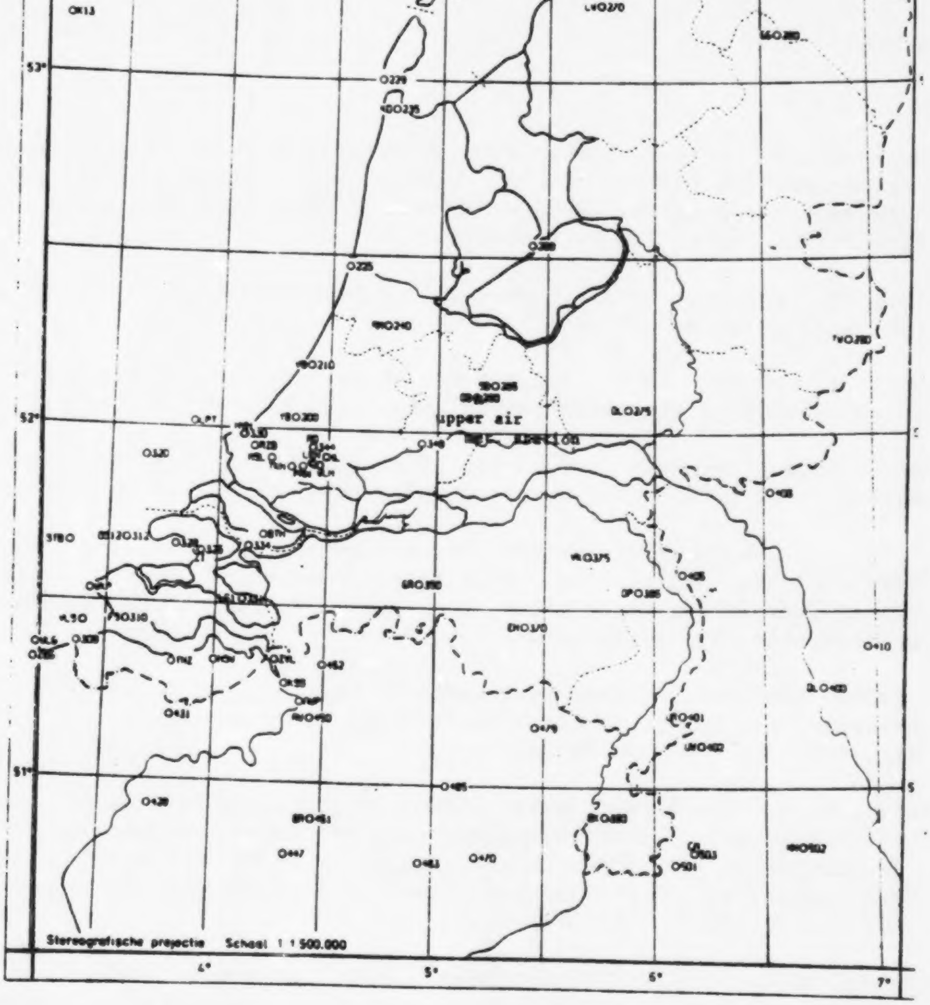
Parameter	Temporal resolution (h)	Frequency	Location[a]	Year of record
In aerosols				
$SO_4^=$	24	daily		1979/1981
NO_3^-	24	daily	1,2,3,4	1981
Cl^-	24	daily		
NH_4^+	24	daily	1,2	1979/1981
Na^+	24	daily	2	1979/1980
H_2SO_4	24	daily	1,2	1979/1980
			3,4	1981
TSP	24	once/6 d		
Carbon	24	once/6 d	1,2	1979/1981
Organic matter in aerosols	24	once/6 d		
HNO_3	24	daily	1,2	1979/1981
			3,4	1981
F^-	24	daily	1	1979/1981
NH_3	24	daily	1	1979/1980
Aldehydes	24	daily	1	1979/1981
Formic acid	24	once/6 d		
Acetic acid	24	once/6 d	1	1979/1981
Propionic acid	24	once/6 d		
Organic compounds				
C_6–C_{18} as gas	1	once/6 d twice/day	1,2,4,5	1979/1981
C_6–C_{18} in aerosol	24	incidental	1	1980/1981
Pesticides	24	once/6 d	1	1979
		once/18 d	1	1980/1981
Scattering coefficient	1	hourly	6	1979/1981
Particle size distribution (EAA)	1	hourly	6	1979/1981

[a] For location, see Table 1.

procedures are implemented for both sampling and analysis, and most of the data are very reliable. Measurements of H_2SO_4, HNO_3, and organic matter in aerosols are less reliable due to possible interference.

Data on surface meteorology in The Netherlands can be obtained from a synoptic meteorological network of 22 stations (Figure 4) covering The Netherlands (Personal communication, Van Dop, 1983). This network is operated by the Royal Netherlands Meteorological Institute, KNMI, and provides hourly standard synoptic observations. One station is part of an upper air radiosonde network. Rawinsondes to measure pressure, temperature, humidity, and wind are released twice a day (at 0000 and 1200 GMT) to a maximum height of 20 km. Pibals for wind measurements are released daily at 0600 and 1800 GMT.

In The Netherlands (as elsewhere), concern is growing over acid deposition. Since 1978, KNMI and RIV (1981) have operated a network of 12 stations, measuring the concentrations in rainwater of several components as well as those of some metals and organic compounds. These components are H^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Zn^{2+}, F^-, Cl^-, NO_3^-, $SO_4^=$, HCO_3^-, and $PO_4^=$. The National Institute for Water Supply, RID, has operated a similar network of 26 stations since 1978 (RID, 1980). Recently, these networks were integrated. However, estimates have shown that at least half of the acid deposition in The Netherlands is dry deposition, and there is an urgent need to develop and apply methods for measuring dry deposition fluxes.


4°
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O102
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O229
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upper air
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O320
O330
O334
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O428
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O501
O502
Stereografische projectie Schaal 1:1 500.000

Figure 4. Surface meterological stations.

ACKNOWLEDGMENTS

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PHOTOCHEMICAL OXIDANTS IN NORTHWESTERN EUROPE, 1976-1979
A PILOT STUDY*

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INTRODUCTION

This pilot study resulted from the growing concern about photochemical air pollution in Europe during the last decade. From other studies conducted in several countries, it had become evident that oxidants and their precursors could be transported over many hundreds of kilometers and that they could affect countries other than those of the precursor sources. In a report from the Organization of Economic Cooperation and Development (OECD, 1978), the Ad Hoc Group of Experts on Photochemical Oxidants and Their Precursors in the Atmosphere concluded that, in view of long-range transport, emission control on a local scale may be grossly insufficient in Europe and Eastern North America (OECD, 1978).

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In 1978, the Norwegian Institute for Air Research (NILU) hosted a planning conference on future cooperative research efforts on the long-range transport of photochemical oxidants (NILU, 1978). Participants from 12 countries in Europe and North America attended the conference, which concluded with several proposals for future research, both on a national basis and in the form of international cooperation. The proposals covered the following major research areas: emissions, transformation, ambient measurements, effects, and integrated modeling studies. The pilot study discussed here, which was conducted as a result of these recommendations, focused on large-scale oxidant episodes in Northwestern Europe during the years 1976 to 1979. The term "pilot study" is used in order to emphasize that the subject has by no means been covered in full detail.

In December 1979, a questionnaire was sent to individuals and institutions in 10 countries in Northwestern Europe, requesting information on measurements of O_3 and other secondary air pollutants for 1976 to 1979. Data were available from eight countries: Austria, Belgium, the Federal Republic of Germany (FRG), Finland, The Netherlands, Norway, Sweden, and the United Kingdom (UK). Requests for more specific data were sent in the spring and summer of 1980.

The formation and transport of photochemical oxidants in northwestern Europe have been frequently discussed in the literature over the last 10 yr. This research has been conducted mainly in the FRG, The Netherlands, the UK, and Scandinavia. Some of the relevant references are Atkins et al. (1972), Cox et al. (1976), Grennfelt (1975, 1976), Fricke and Rudolf (1977), Guicherit and van Dop (1977), Apling et al. (1977), Guicherit (1978), Becker et al. (1979), and Schjoldager (1979, 1980). A relatively detailed literature survey is provided in the project report for the pilot study (Schjoldager et al., 1981).

OZONE MONITORING STATIONS

Eight countries provided information on their O_3 monitoring stations for the pilot study. These stations are described below. Included in each description is information on the type of station, location of the station (altitude, latitude, longitude), period for which the data were collected, the measurement method, the calibration method, and additional comments regarding the availability of data on secondary pollutants.

Station: IMP, Vienna, Austria

Type:	Urban, 1 m above street level.
Altitude:	180 m.
Coordinates:	Latitude 48°13' N, longitude 16°22' E.
Period:	July-September 1976; March-September 1977; 1978 and 1979.
Measurement:	Chemiluminescence (ethylene).
Calibration:	KI, EPA (Federal Register, 1971).
Comments:	HC and NO_x measurements also available.

Station: AfL, Vienna, Austria

Type:	Urban, 14 m above street level.
Altitude:	193 m.
Coordinates:	Latitude 48°13' N, longitude 16°22' E.
Period:	March-July 1976, May-September 1977, March-July 1978, March-September 1979.
Measurement:	Chemiluminescence (ethylene).
Calibration:	KI, EPA (Federal Register, 1971).
Comments:	HC and NO_x measurements also available.

Station: Illmitz, Austria

Type: Rural, 65 km southeast of Vienna.
Altitude: 119 m.
Coordinates: Latitude 47°46' N, longitude 16°46' E.
Period: May-September 1978, April-September 1979.
Measurement: Chemiluminescence (ethylene).
Calibration: KI, EPA (Federal Register, 1971).
Comments: NO_x measurements also available.

Station: Roeschitz, Austria

Type: Rural, 65 km northwest of Vienna.
Altitude: 282 m.
Coordinates: Latitude 48°40' N, longitude 15°53' E.
Period: April-September 1979.
Measurement: Chemiluminescence (ethylene).
Calibration: KI, EPA (Federal Register, 1971).
Comments: NO_x measurements also available.

Station: R 822, Antwerp area, Belgium

Type: Suburban/industrial, 3 m above surface.
Altitude: 8 m.
Coordinates: Latitude 51°16' N, longitude 4°22' E.
Period: March-September 1979.
Measurement: Chemiluminescence.

Station: R 801, Antwerp area, Belgium

Type: Urban.
Coordinates: Latitude 51°13' N, longitude 4°26' E.
Period: March-September 1979.
Measurement: Chemiluminescence.
Comments: HC and NO_x data available from 13 and 18 Belgian stations, respectively.

Station: Zentralstation, Frankfurt, FRG

Type: Urban.
Period: June-August 1976, June-July 1977, June-August 1978, May-July 1979.
Measurement: Chemiluminescence (ethylene).
Calibration: UV/absorption.

Station: Feldberg, Frankfurt, FRG

Type: Rural, mountain station.
Altitude: 805 m.
Period: March-September 1976, 1977, and 1978.
Measurement: Chemiluminescence.
Calibration: UV/absorption.

Station: Venusberg, Bonn, FRG

Type: Suburban.
Altitude: 220 m.
Period: March-September 1976, 1977, and 1978.
Measurement: Chemiluminescence.
Calibration: UV/absorption.
Comments: O_3 data from five other stations in the Cologne-Bonn area (Eifelwall, Godorf, Bonn Universitat, Olberg, and Michelsberg); NO_x and HC data available from several stations.

Station: Helsinki, Finland

Type: Urban.
Coordinates: Latitude 60° N, longitude 25° E.
Period: April-August 1979.
Measurement: Chemiluminescence (rhodamine B).

Station: Delft, Netherlands

Type: Suburban.
Altitude: 1.5 m.
Coordinates: Latitude 52°00' N, longitude 4°23' E.
Period: March-September 1976, 1977, 1978, and 1979.
Measurement: Galvanometric (1976, 1977), colorimetric (1978), chemiluminescence, ethylene (1979).
Calibration: Electrochemistry (1976), gas-phase titration (1977 to 1979).

Station: Terschelling, Netherlands

Type: Rural.
Altitude: 4 m.
Coordinates: Latitude 53°24' N, longitude 5°21' E.
Period: June-August 1978.
Measurement: Chemiluminescence (ethylene).
Calibration: Gas-phase titration.
Comments: O_3, HC, and NO_x data available from many other stations in the Netherlands. Measurements of peroxyacetyl nitrate (PAN) and peroxybenzoyl nitrate (PBzN) have also been performed (Guicherit, 1978).

Station: Maridalen, Oslo, Norway

Type:	Rural, 15 km north of Oslo.
Altitude:	165 m.
Coordinates:	Latitude 60°00' N, longitude 10°48' E.
Period:	May-September 1977, June-September 1978, May-September 1979.
Measurement:	Chemiluminescence (ethylene).
Calibration:	KI, EPA (Federal Register, 1971).

Station: Bjornstad, Telemark, Norway

Type:	Suburban/industrial.
Altitude:	30 m.
Coordinates:	Latitude 59°09' N, longitude 9°38' E.
Period:	May-September 1976, May-August 1977, May-September 1978.
Mesurement:	Chemiluminescence (rhodamine B).
Calibration:	Same as Maridalen.

Station: Langesund, Telemark, Norway

Type:	Suburban/coastal.
Altitude:	10 m.
Coordinates:	Latitude 59°01' N, longitude 9°45' E.
Period:	April-September 1979.
Measurement:	Chemiluminescence (rhodamine B).
Calibration:	Same as Maridalen.

Station: Haukenes, Telemark, Norway

Type:	Rural.
Altitude:	30 m.
Coordinates:	Latitude 59°12' N, longitude 9°29' E.
Period:	April-September 1979.
Measurement:	Chemiluminescence (rhodamine B).
Calibration:	Same as Maridalen.
Comments:	O_3 data from three other stations available for 1978 to 1979. HC and NO_x data available from some of the Telemark stations.

Station: Roervik, Sweden

Type:	Rural/coastal.
Altitude:	20 m.
Coordinates:	Latitude 57°25' N, longitude 11°56' E.
Period:	May-September 1976, 1977, 1978, and 1979.
Measurement:	Chemiluminescence (ethylene).
Calibration:	KI, EPA (Federal Register, 1971).
Comments:	NO_x data also available.

Station: Goeteborg, Sweden

Type: Urban, 20 m above street level.
Altitude: 25 m.
Coordinates: Latitude 57°43' N, longitude 12°00' E.
Period: May-September 1976, 1977, 1978, and 1979.
Measurement: Chemiluminescence (ethylene).
Calibration: Same as Roervik.
Comments: HC and NO_x data also available. O_3 data from two stations on the Swedish east coast (Stockholm and Mollergren, 1978).

Station: WSL, UK

Type: Suburban/rural.
Altitude: 100 m.
Coordinates: Latitude 51°53' N, longitude 00°12' W.
Period: April-September 1977, March-September for 1978 and 1979.
Measurement: Chemiluminescence (ethylene).
Calibration: Neutral buffered KI, cross-referenced with UV/absorption.

Station: London, UK

Type: Urban.
Altitude: 6 m.
Coordinates: Latitude 51°29' N, longitude 00°08' W.
Period: March-September 1976, March-September 1977, March-September 1978, March-September 1979.
Measurement: Same as WSL.
Calibration: Same as WSL.

Station: Islington, UK

Type: Urban.
Altitude: 20 m.
Coordinates: Latitude 51°32' N, longitude 00°06' W.
Period: March-September for 1976, 1977, 1978, and 1979.
Measurement: Same as WSL.
Calibration: Same as WSL.

Station: Sibton, UK

Type: Rural.
Altitude: 46 m.
Coordinates: Latitude 52°18' N, longitude 01°28' E.
Period: July-September 1976, March-September 1977, March-September 1978, April-September 1979.
Measurement: Same as WSL.
Calibration: Same as WSL.

Station: Canvey, UK

Type: Suburban/rural.
Altitude: 3 m
Coordinates: Latitude 51°32' N, longitude 00°34' E.
Period: May-September 1977, March-September 1978, and 1979.
Measurement: Same as WSL.
Calibration: Same as WSL.

Station: Harrow, UK

Type: Suburban.
Altitude: 60 m.
Coordinates: Latitude 51°34' N, longitude 00°21' W.
Period: August-September 1979.
Measurement: Same as WSL.
Calibration: Same as WSL.

In addition to the six UK stations listed above, four other stations provided data for the oxidant episode occurring June to July 1976: MRC City, GLC County Hall, GLC Hainault, and GLC Teddington (Ball and Bernard, 1978). Also, data from Harwell were made available for certain episodes in 1977 and 1978, and some data from Lancaster for 1977 and 1978 are available in the literature (Harrison and Holman, 1979; Harrison and McCartney, 1980). Harwell is a rural site with an altitude of 130 m, a latitude of 51°34' N, and a longitude of 1°19' W.

SUMMARY OF OZONE MEASUREMENTS

High Concentrations of Ozone

Many factors make a comparison from year to year or from country to country difficult. Both the number of stations and the type (urban, suburban, rural) vary from year to year. Furthermore, the meteorological conditions favorable for oxidant formation vary considerably from one year to the next. Finally, the calibration methods used to collect data are not consistent.

Data on concentrations ≥200 ppb are given in Table 1. The "Grosswetterlagen" (GWL) categories, which are daily categories published by the FRG's Meteorological Service, are also given.

Data on maximum hourly concentrations exceeding the reference values of 100 ppb and 150 ppb are given on a country basis for the period May to August each year in Table 2. High concentrations during the warm, dry summer of 1976 are evident, as well as the large number of high concentrations in Austria in 1979. As mentioned previously, a comparison from year to year or from country to country should not be made because of inconsistencies in the data base. The number of days with high concentrations may have been underestimated for The Netherlands and the FRG, because these countries collected more O_3 data than those discussed in this paper.

Covariation with the Large-Scale Weather Pattern, GWL

In Table 3, the number of days with maximum hourly O_3 concentrations ≥100 ppb is listed, as well as the total number of days for each GWL category. The English data are grouped under "Great Britain"; the Belgian, Dutch, and German data are grouped under "European Continent"; and the Norwegian and Swedish data are grouped under "Scandinavia". The Austrian data are not included in Table 3.

TABLE 1. HOURLY OZONE CONCENTRATIONS ≥200 ppb.

Station	Date	Maximum Concentration (ppb)	Hours with Concentration ≥200 ppb	GWL[a]
Illmitz, Austria	04-15-79	205	3	U
	06-07-79	220	2	BM
	06-11-79	213	2	BM
	08-15-79	249	3	HFA
	08-22-79	203	2	TRW
	09-14-79	208	2	NWA
Venusberg, Bonn, FRG	07-12-77	202	1	HNA
Delft, Neth.	1976	200		
Vlaardingen, Neth.	05-08-76	270		SEA
Vlissingen, Neth.	06-25-76	208	1	HM
Haamstede, Neth.	07-03-76	261	1	HNA
WSL, UK	07-03-76	207	2	HNA
Harwell, UK	07-02-76	>220	6	BM
	07-03-76	>220	7	HNA
	07-04-76	230	4	HNA
	07-05-76	258	6	HNA
	07-06-76	204	1	HNA
	07-07-76	212	2	HNA
MRC City, UK	06-25-76	201	2	HM
	06-26-76	203	2	HM
	06-27-76	200	1	HM
	07-03-76	>200	1	HNA
GLC Teddington, UK	06-28-76	211	1	HM

[a] Grosswetterlagen.

TABLE 2. OZONE CONCENTRATIONS $\geq$100 ppb AND 150 ppb, MAY TO AUGUST 1976-1979

Country	Reference Value (ppb)	1976	1977	1978	1979
Austria	100	5	15	30	115
	150	0	2	1	65
Belgium	100				6
	150				1
FRG	100	31	6	9	1
	150	6	2	1	0
Finland	100				1
	150				0
Netherlands	100	29	2	3	7
	150	5	0	0	0
Norway	100	6	1	3	4
	150	0	0	0	2
Sweden	100	12	2	14	7
	150	0	0	0	2
UK	100	28	13	7	3
	150	14	1	0	1

TABLE 3. MAXIMUM 1-h O_3 CONCENTRATIONS ≥100 ppb FOR VARIOUS LARGE-SCALE WEATHER PATTERNS, MAY-AUGUST, 1976-79

Description	Weather	EC[a]	GB[b]	SC[c]	Total Days
Grosswetterlagen der zonalen Zirkulationsform					
Westlage, antizyklonal	WA	1		1	27
Westlage, zyklonal	WZ	3		1	61
Suedliche Westlage	WS				6
Winkelfoermige Westlage	WW	1		1	7
Grosswetterlagen der gemischten Zirkulationsform					
Suedwestlage, antizyklonal	SWA				4
Suedwestlage, zyklonl	SWZ				4
Nordwestlage, antizlyklonal	NWA				11
Nordwestlage, zyklonal	NWZ				19
Hoch ueber Mitteleuropa	HM	16	16	8	27
Hochdruckbrucke (Ruecken) ueber Mitteleuropa	BM	16	8	1	54
Tief Mitteleuropa	TM			1	15
Grosswetterlagen der meridionalen Zirkulationsform					
Nordlage, antizyklonal	NA	1			8
Nordlage, zyklonal	NZ				16
Hoch Nordmeer-Island, antizyklonal	HNA	10	10		23
Hoch Nordmeer-Island, zyklonal	HNZ		1		17
Hoch Britische Inseln	HB		1	2	16
Trog Mitteleuropa	TRM				17
Nordostlage, antizyklonal	NEA	2	2	1	22
Nordostlage, zyklonal	NEZ	1	2	1	26

(continued)

TABLE 3. (continued)

Description	Weather	EC[a]	GB[b]	SC[c]	Total Days
Hoch Fennoskandien, antizyklonal	HFA	8	3	5	24
Hoch Fennoskandien, zyklonal	HFZ				13
Hoch Nordmeer-Fennoskandien, antizyklonal	HNFA	4	2	4	12
Hoch Nordmeer-Fennoskandien, zyklonal	HNFZ		1	2	11
Suedostlage, antizyklonal	SEA	3	3	1	4
Suedostlage, zyklonal	SEZ				
Suedlage, antizyklonal	SA				
Suedlage, zyklonal	SZ			2	2
Tief Britische Inseln	TB			2	13
Trog Westeruopa	TRW	1	1	4	34
Uebergang	U	2	1	1	3
Total		69	51	38	492

[a] EC = European Continent (Belgium, Netherlands, Federal Republic of Germany).
[b] GB = Great Britain (United Kingdom).
[c] SC = Scandinavia (Norway, Sweden).

Although high O_3 levels were strongly associated with some weather events (e.g., HM, BM, HNA, HFA and HNFA), most of these events did not necessarily imply high O_3 concentrations. Thus, it seems that large-scale weather patterns alone do not determine conditions sufficient for O_3 formation. An exception to this is the HM category, showing high O_3 concentrations in UK and in Europe during 16 of 27 days.

Certain weather patterns are grouped together in Table 4 in order to examine differences between Scandinavia and the rest of Europe. The relative occurrence of high O_3 levels for the categories HM, HFA, and HNFA was similar for the three regions. For the categories BM, HNA, and SEA, there were many high values for Europe and the UK, but there were considerably fewer high values for Scandinavia. The opposite was the case for the categories HNFZ, SZ, TB, and TRW; there were few high values for Europe and the UK and high values for Scandinavia. Some of these categories may be associated with pollutant transport to Scandinavia from other parts of Europe.

TABLE 4. MAXIMUM 1-h O_3 CONCENTRATION >100 ppb FOR VARIOUS LARGE-SCALE WEATHER PATTERN CATEGORIES, MAY-AUGUST, 1976-1979

	EC[a]		GB[b]		SC[c]	
Category	(No.)	(%)	(No.)	(%)	(No.)	(%)
HM, HFA, HNFA	28	41	21	41	17	45
BM, HNA, SEA	29	42	21	41	2	5
HNFZ, SZ, TB, TRW	1	1	2	4	10	26
Other	11	16	7	14	9	24
Total	69	100	51	100	38	100

[a] EC = European Continent (Belgium, Netherlands, Federal Republic of Germany).

[b] GB = Great Britain (United Kingdom).

[c] SC = Scadinavia (Norway, Sweden).

SELECTED EPISODES

This section contains a discussion of selected time periods during which the O_3 concentrations exceeded 100 ppb at several stations. The discussion is based on daily weather maps, including synoptic weather situations and local meteorological conditions, and on 850-mbar air trajectories. The trajectories were calculated as part of OECD's study, Long-Range Transport of Air Pollutants (LRTAP) in 1976 and 1977 (OECD, 1977) and as part of ECE's study, European Monitoring and Evaluation Programme (EMEP) in 1978 and 1979 (ECE, 1977). The 850-mbar trajectories should not be used to identify definite precursor source regions, but as rough indicators of the air flow aloft, especially during high pressure situations when the trajectories are more uncertain.

This paper presents information on the following two episodes, June 19 to July 17, 1976, and May 30 to June 8, 1979. In the project report, six other episodes are also discussed (Schjoldager et al., 1981):

- August 16 to August 30, 1976,
- June 12 to June 15, 1977,
- July 2 to July 12, 1977,
- July 28 to August 1, 1978,
- August 20 to August 23, 1978, and
- May 12 to May 20, 1979.

For each of the selected episodes, the daily (1200 GMT) weather maps are given for every second day. The weather maps are from Weather Log, published by the British Meteorological Office. Air trajectories at the 850-mbar level are presented. For 1976 and 1977, 48-h trajectories are available; for 1978 and 1979, 96-h trajectories are available.

June 19 to July 17, 1976, Episode

This episode, which was discussed in the literature by Apling et al. (1977) and Ball and Bernard (1978), was characterized by high pressure centers over various parts of Europe with abnormally warm and dry weather. According to Weather Log (1976), the hot spell in the UK was "probably unprecedented in length and intensity since the eighteenth century."

The weather maps for every second day of the period are given in Figure 1. At the beginning of the period, the high pressure center moved eastward from the Atlantic Ocean, covering large parts of central Europe, while a low pressure area was located south of Iceland. The high pressure center later moved slowly towards the Norwegian Sea. During most of the period, wind speed was low and maximum temperatures exceeded 25°C. Wind direction was often variable. The skies were mostly clear, except for the first and last days in the period.

In Figure 2, the 48-h air trajectories at the 850-mbar level, arriving at 1200 GMT on every second day, are presented. The air trajectories indicate transport aloft from the west during the first days of the period. Towards the end of June, the transport to Scandinavia and the UK was from the southwest, while there was variable transport on the continent. In the beginning of July, the air aloft moved clockwise around the high pressure center in the North Sea.

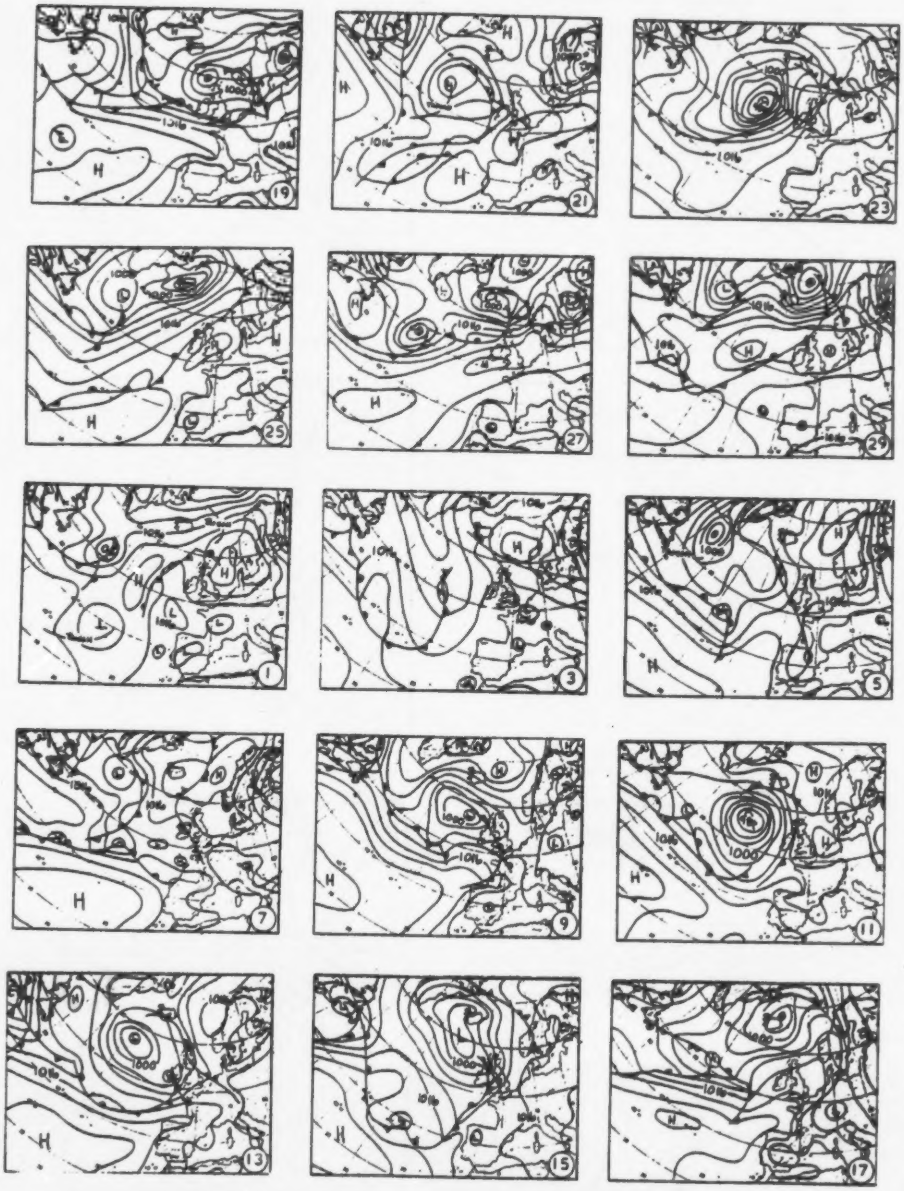
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Figure 1. Daily weather maps at 1200 GMT for every second day, June 19-July 17, 1976 (British Meteorological Office, 1976).

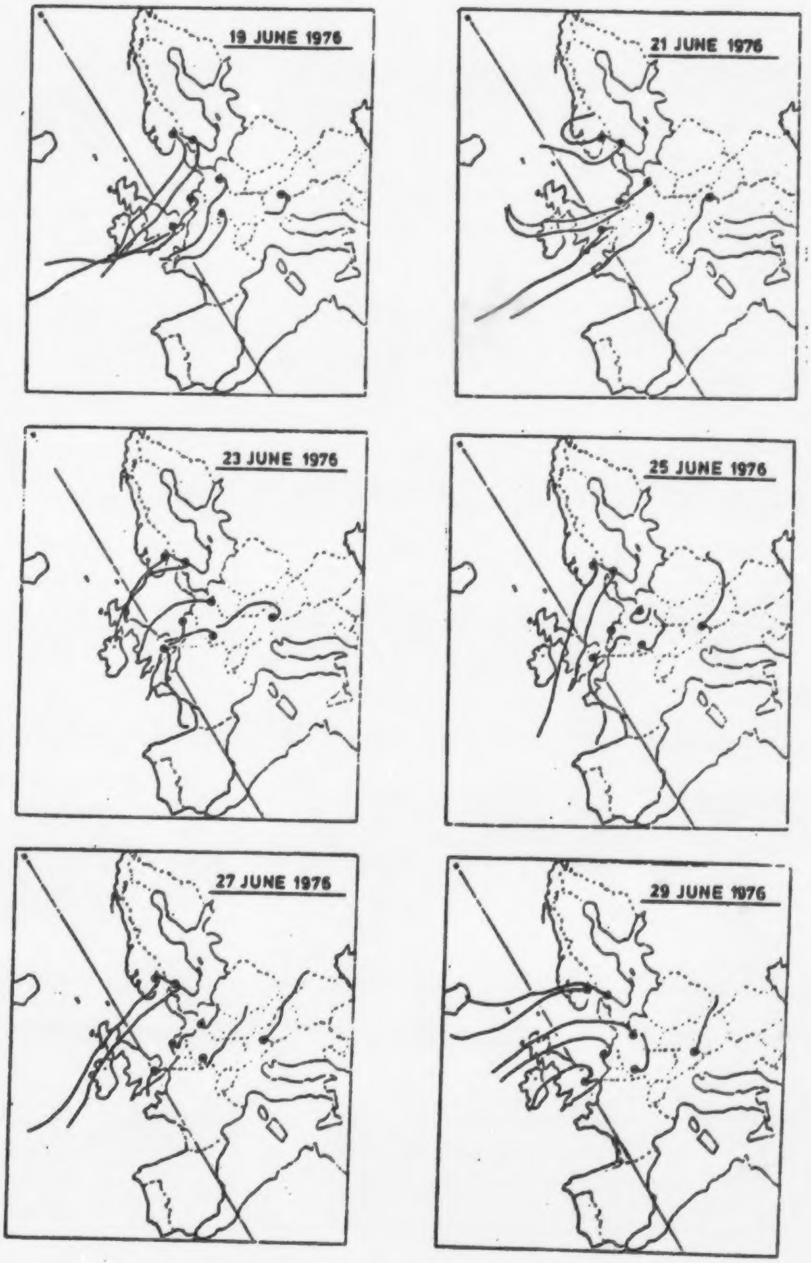
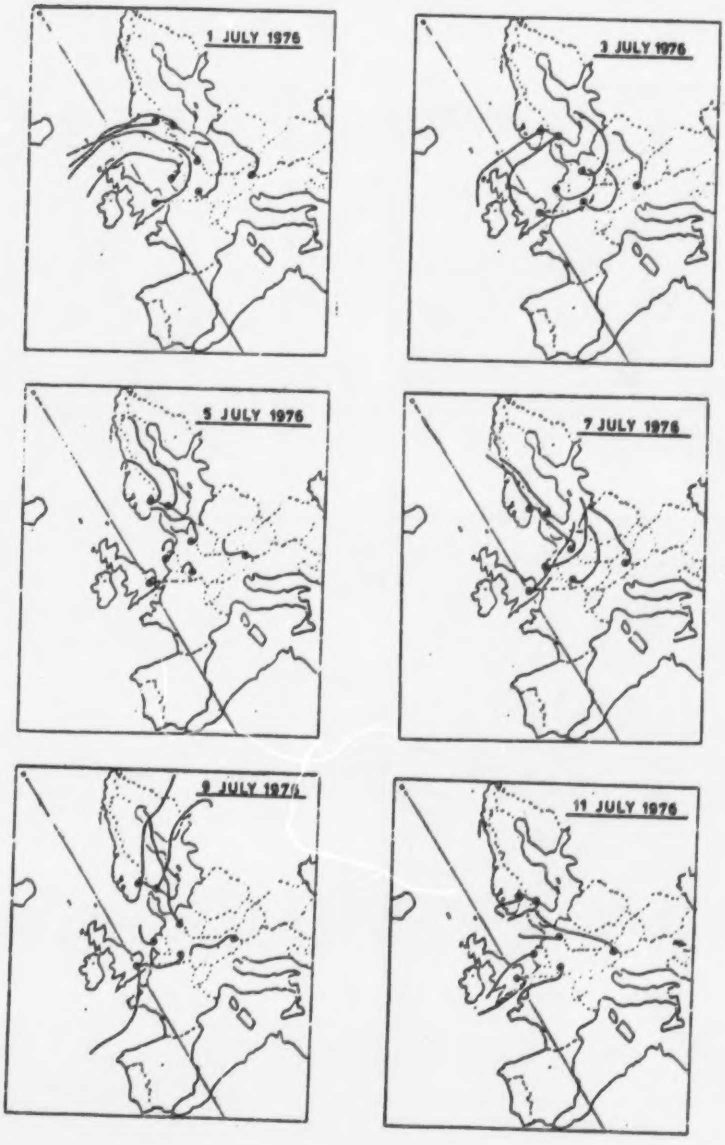

19 JUNE 1976
21 JUNE 1976
23 JUNE 1976
25 JUNE 1976
27 JUNE 1976
29 JUNE 1976

Figure 2. The 48-h air trajectories at the 850-mbar level arriving at 1200 GMT on every second day, June 19-July 17, 1976.


1 JULY 1976
3 JULY 1976
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9 JULY 1976
11 JULY 1976

Figure 2. (continued).

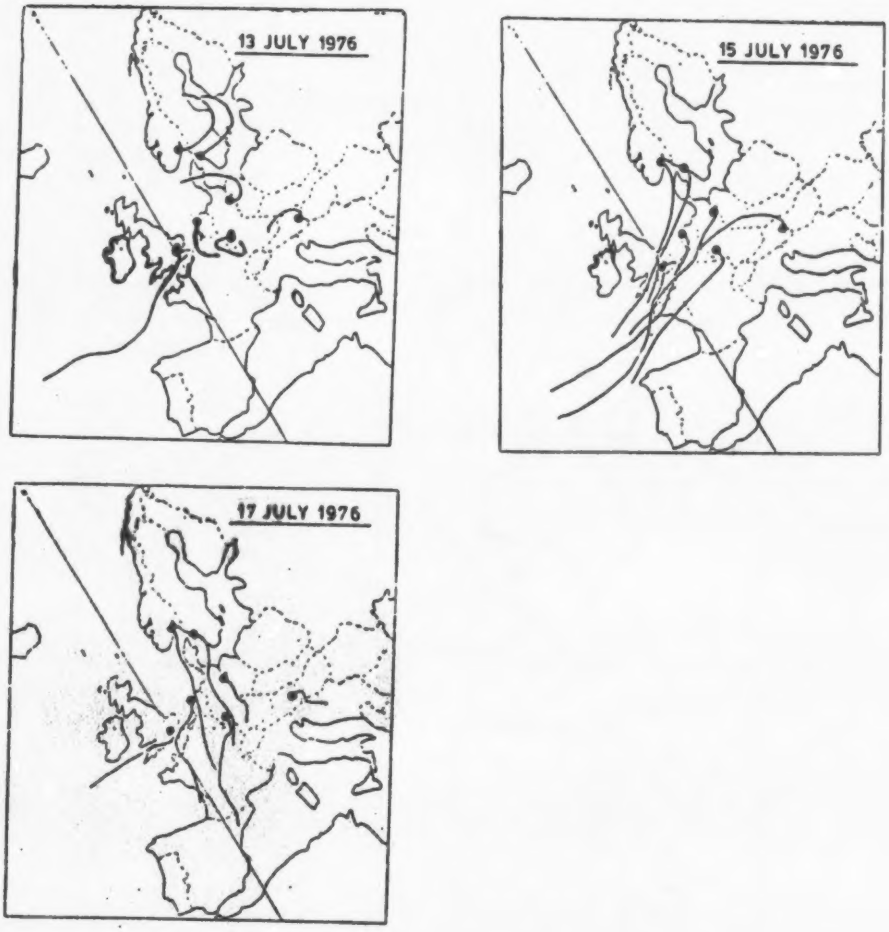
13 JULY 1976
15 JULY 1976
17 JULY 1976

Figure 2. (continued).

Between July 5 and July 15, there was generally no large-scale transport aloft; this lasted until the end of the period when a cold front approached from the Atlantic Ocean.

The maximum hourly concentrations are given on a daily basis in Table 5. The O_3 concentrations were high over all of Europe, reaching 129 ppb in Austria, 186 ppb in Germany, 191 ppb in The Netherlands, 258 ppb in Great Britain, and 125 ppb in Sweden.

As explained earlier, the highest concentrations in the UK resulted from a combination of O_3 transport from the European continent and local and mesoscale formation (Apling et al., 1977; Ball and Bernard, 1978). This explanation may in general be valid for other countries as well. During the long-lasting high pressure situation, transboundary air pollution probably affected large parts of Europe, interacting with the locally emitted oxidant precursors.

May 30 to June 8, 1979, Episode

An anticyclone stagnated over Scandinavia during most of the period, and the pressure gradients over Europe were generally small (Figure 3). The local weather was fair in southern Scandinavia and on the continent. In England, it was much more cloudy.

The air trajectories indicate transport from the south to Western Europe in the beginning. Later, the transport had a significant easterly component, changing to west at the end of the period (Figure 4).

TABLE 5. MAXIMUM HOURLY O_3 CONCENTRATIONS (ppb) AND LARGE-SCALE WEATHER PATTERN (GWL), JUNE 19-JULY 17, 1976

Station	June											
	19	20	21	22	23	24	25	26	27	28	29	30
Alf, Wien	119	79	63	63	78	50	80	95	84	129	89	68
Zentralstation, Frankfurt	98	63	116	186	87	127				121	124	95
Feldberg, Frankfurt	92	57	63	89	82		83	48	46	53	54	38
Venusberg, Bonn	52	36	45	104	70	75	82	70	70	101	75	61
Delft, Netherlands	45	36	64	119	119	191	151	119	154	96	46	69
Vlissingen, Netherlands	52	90	59	89	152	128	208	176	176	168	98	140
Harwell, England				149	116	122	162				160	111
WSL, England				64	101	83	115	139	149	143	97	75
MRC City, England				60	72	96	201	203	200	138	78	75
Roervick, Sweden	96	90	59	57	74	73	119	114	97	58	99	70
Bjoernstad, Norway	69	55	70	62	42		70	101	84	54	55	69
GWL	WZ	WZ	WZ	HM	HM	HM	HM	HM	HM	HM	BM	BM

(continued)

TABLE 5. (continued)

Station	July																
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Alf, Wien	72	61	103	93	100	48		72	83	43	50						
Zentralstation, Frankfurt	110	116	160	171	173	77	100	174	103	93	112	143	48	110	94	119	152
Feldberg, Frankfurt		101	148	138	122	78	89	140	98	86							
Venusberg, Bonn	62	73	94	130	115	76	82	170	73	48	79	86	38	47	45	64	65
Delft, Netherlands	47											68	42	43	86	95	57
Vlissingen, Netherlands	108	158	261	148	140	142	140	96	68	73	88	112	68	49	70	112	72
Harwell, England	172	>220	>220	230	258	204	212	196	62	72	118	171					
WSL, England	117	125	207	152	138	164	118	139	57	76	122	164					
MRC City, England	96	169	>200	156	106	125	104	111	34	73	81	99					
Roervick, Sweden	80	87	73	89	68	97	85	71	49	73	64	73	93	107	96	125	93
Bjoernstad, Norway	48	89				93	104	67	57	78	67	70	70	86	85	104	73
GWL	BM	BM	BM	HNA	HNA	HNA	HNA	HNFA	HNFA	HNFA	HNFA	HFA	HFA	HFA	HFA	HNA	BM

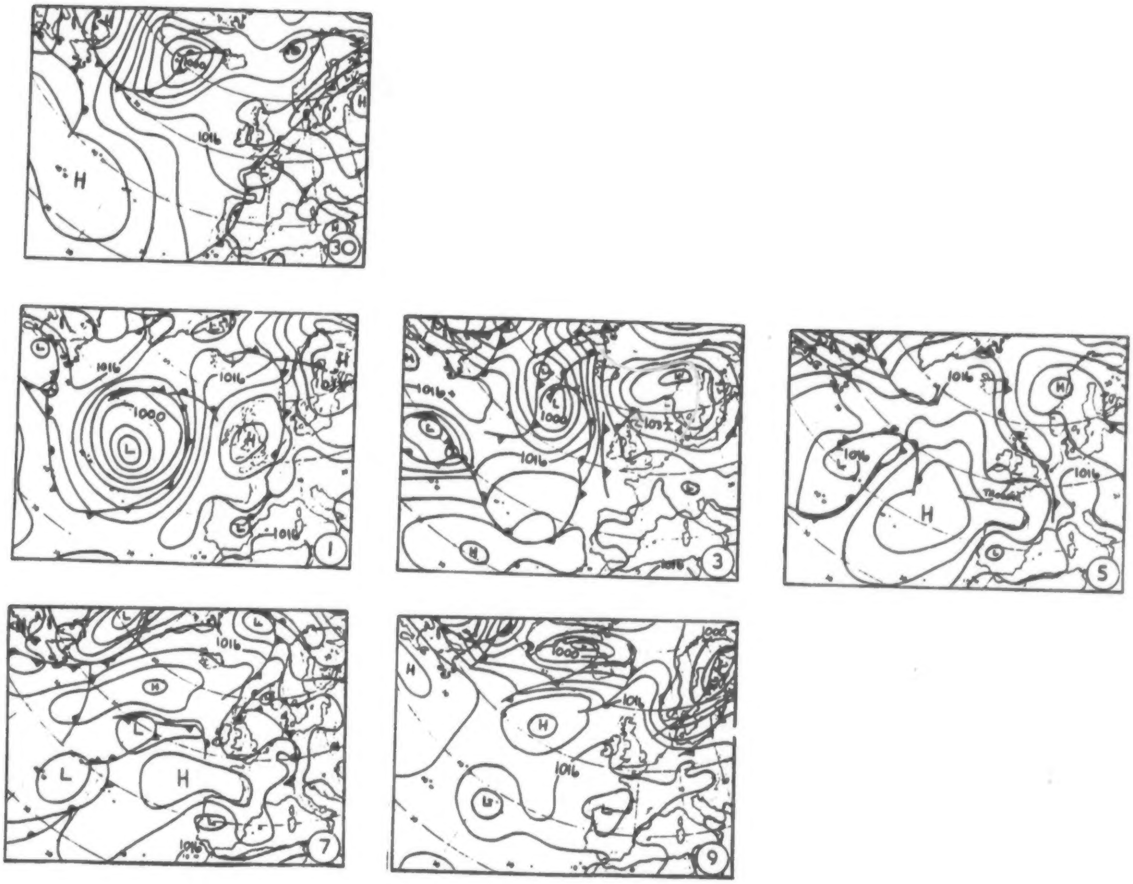
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Figure 3. Daily weather maps at 1200 GMT for every second day, May 30-June 1979 (British Meteorological Office, 1979).

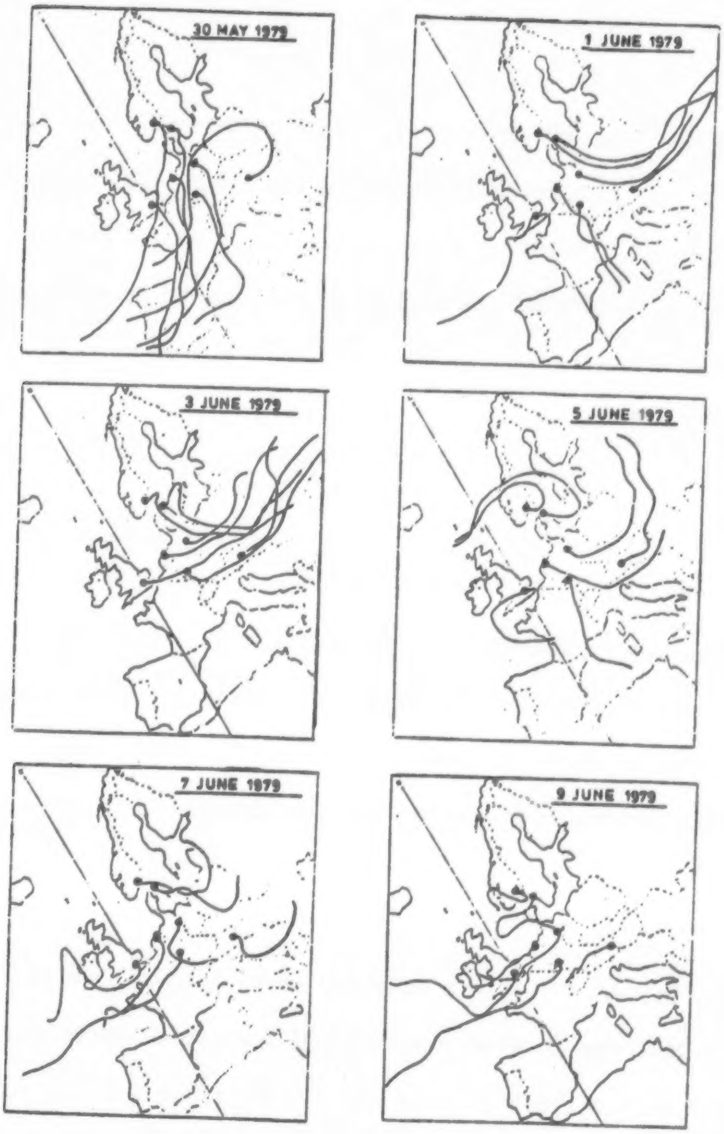
30 MAY 1979
1 JUNE 1979
3 JUNE 1979
5 JUNE 1979
7 JUNE 1979
9 JUNE 1979

Figure 4. The 96-h air trajectories at the 850-mbar level, arriving at 1200 GMT on every second day, May 30-June 9, 1979.

The O_3 concentrations were high in Austria and low in England throughout the period (Table 6). Maximum concentrations exceeding 100 ppb also occurred in Belgium and the FRG. In Sweden, the maximum hourly concentrations were 80 to 50 ppb. In Norway, the maximum concentration of 197 ppb occurred downwind of an industrial area (Schjoldager, 1980).

It may be the high concentrations in Central Europe were due to local and mesoscale formation with some enhancement from long-range transport. For southern Scandinavia, transport from distant sources was probably important in the first part of the period, but local and mesoscale production appear to have contributed significantly during last part of the period.

CONCLUSIONS

With one exception, all the hourly O_3 concentrations exceeding 200 ppb were measured in England or The Netherlands during 1976, or at Illmitz, Austria, during 1979. The highest 1-h O_3 concentration discussed in this report is 258 ppb, measured at Harwell, England, on July 5, 1976. However, concentrations up to 0.27 ppm were reported earlier at Vlaardingen, Netherlands, on May 8, 1976.

Most high O_3 concentrations occurred with stagnating anticyclones. When a high pressure area was located over Central Europe, Scandinavia, or Finland, the concentrations were often high over all the examined regions in Northwestern Europe. With a high pressure ridge over Central Europe or with the high pressure center over the North Sea, the concentrations could be high in Europe

TABLE 6. MAXIMUM HOURLY O_3 CONCENTRATIONS (ppb) AND LARGE-SCALE WEATHER PATTERNS (GWL), MAY 30-JUNE 8, 1979

	May		June							
Station	30	31	1	2	3	4	5	6	7	8
Alf, Wien	92	100	108	119	119	128	99	154	160	70
Illnitz, Austria	161	183	189	186	198	193	180	166	220	161
Roschnitz, Austria	107	111	99	98						84
Zentralstation, Frankfurt	50	122	48	66	63	78	70	23	42	30
Delft, Netherlands	28	56	53	40	66	78	47	48	41	65
Canvey, UK	45	23	58	41	65	65	44	42	30	41
WSL, UK	40		29	27	25	56	25	23	17	28
Sibton, UK	56	43	53	44	77	75	61	47	43	60
Rorvik, Sweden	114	112	150	114	80	95	90	92	84	
Langesund, Norway	57	94	90	70	61	65	71	93	90	64
R 801, Belgium										
R 822, Belgium		47	44	39	100	63		37		28
Helsinki, Finland	63	56	44	57	50	51	54	57	57	59
GWL	HNFA	HNFA	HNFA	HNFA	HNFA	HNFA	HNFA	HNFA	BM	BM

and in Great Britain but significantly lower in Scandinavia. In cases with cyclonic circulations and low pressure areas over Central Europe, the concentrations could remain high in Scandinavia while they were significantly lower in other parts of Northwestern Europe.

At the Austrian station of Illmitz, located in a rural area 65 km southeast of Vienna, the concentrations were high throughout the period April to September 1979. During this period, the maximum concentration exceeded 150 ppb on 90 days.

Two episodes were studied in more detail with respect to synoptic weather situations, local meteorological conditions, and trajectory analyses, in order to assess the origin and transport of the polluted air masses. Six other episodes are discussed in the project report of the pilot study. In some cases, long-range transport appeared to be more important than local and mesoscale O_3 formation, while the contrary occurred on other occasions. Due to the methodology used and the limited measurement data, it was impossible to assess quantitatively the role of various production scales. Thus, it has not been possible to evaluate quantitatively the influence of the various precursor source regions on the O_3 concentrations at the various receptor points.

The highest O_3 concentrations were reached during weather conditions conducive to oxidant formation, when local precursors were emitted into polluted air masses transported from other source regions. A typical episode of this kind occurred in England during the hot spell in June/July 1976.

In most parts of Northwestern Europe, the maximum O_3 concentrations are as high as, and in some cases higher than, the threshold levels associated with plant damage and health effects.

The need for further, concerted studies of photochemical oxidants in Europe is clearly present.

RECOMMENDATIONS

Among the many aspects of photochemical air pollution in Europe that should be investigated further are the following:

- An emissions inventory of NO_x and VOCs for northwestern Europe. The emissions should be given for grid squares of approximately 100 km x 100 km. Volatile organics should be specified in terms of chemical reactivity.
- The transport of air pollutants during high pressure situations. In these cases, air trajectories are often highly uncertain. The highest concentrations of photochemical oxidants are, however, most likely to occur during these weather situations. The need for more research on this issue is thus obvious.
- Large-scale photochemical transport models. These models will increase the general understanding of and aid in predicting the effect of future emission controls.
- A consistent data base of relevant air concentrations. Consistency in measurement and calibration methods, and in the criteria for sampling sites is essential.
- Measurements above the surface layer. Aircraft measurements are of value in determining the horizontal and vertical extent of high concentrations. Measurements from meteorological towers are of value in the study of the diurnal concentration variation above the nocturnal surface inversion layer.

It should also be pointed out that other parts of Europe may experience concentration levels of photochemical oxidants as high or higher than those given in this report. Of special importance is the entire southern part of Europe. The total precursor emissions there are smaller than those measured in Northwestern Europe, but the weather of Southern Europe is much more sunny and warm.

It may turn out that in all the Mediterranean countries, from Spain to Turkey, oxidant levels exceeding internationally accepted threshold values can occur. Studies in these regions are thus highly desirable.

The Austrian O_3 data also indicate that high concentrations can occur in parts of Central Europe. Measurements downwind of other major metropolitan areas of Central Europe will give insight into this phenomenon.

In order to study large-scale photochemical oxidant formation and transport in Northwestern Europe without local influence, rural monitoring sites are preferred. For the establishment of a minimum sampling network in northwestern Europe, a distance of about 300 km between neighbouring stations is recommended. Table 7 lists the number of stations that would be required in each country to meet such a distribution.

TABLE 7. NUMBER OF STATIONS PROPOSED FOR A MINIMUM SAMPLING NETWORK IN NORTHWESTERN EUROPE

Country	No. of Stations
Austria	2
Belgium	1
Denmark	1-2
FRG	3
Finland	2
France	2-3
Ireland	1
Netherlands	1
Norway	2-3
Sweden	2-3
UK	2-3

The air pollutants of interest may tentatively be grouped into priority categories as follows:

First priority: O_3 (continuous)

Second priority: Sulphate (24-h)

Visibility (at least once per day)

Nitric acid (24-h)

PAN (continuous, if possible)

Third priority: NO_x (continuous)

VOC (continuous)

SO_2 (24-h)

The quality of the measurement data should be assured by regular intercalibration procedures and station performance audits.

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FRG: Ulrich Schurath, Institut for Physikalische Chemie der Universitat Bonn, Bonn; H-W Georgii, Institut fur Meteorologie und Geophysik der Johann Wolfgang Goethe-Universitat, Frankfurt a.M; Werner Rudolf, Umweltbundesamt Pilotstation Frankfurt, Frankfurt a.M.

Finland: Risto Lahdes, Helsingin kaumpungin terveysvirasto, Helsinki.

Netherlands: Robert Guicherit, Research Institute for Environmental Hygiene TNO, Delft.

Norway: Leif Stige, Norwegian State Pollution Control Authority (SFT), Porsgrunn.

UK: Richard G. Derwent, Environmental and Medical Sciences Division, AERE Harwell, Oxfordshire; Alan J. Aplig, Warren Spring Laboratory, Department of Industry, Gunnels Wood Road, Stevenage.

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Emissions Inventories in Europe*

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INTRODUCTION

The most important objective of an air pollution control policy is the maintenance of air quality, a goal affecting humans, animals, and the biological and material environment. In addition to the effects of air pollutants in the neighborhood of air pollution sources, the effects of the long-range transport of these pollutants are also important. Sulfur compounds and nitrogen compounds are mainly responsible for the presence of oxidant pollutants.

An important element of an air quality management system is the establishment and implementation of clean air plans, and the basis for any clean air plan is an emissions inventory. According to the German Clean Air Act, clean air plans and emissions inventories must be established in heavily polluted areas (HPAs), which must be declared as such by the German provinces. Other European countries like The Netherlands, Belgium, Denmark and Norway are also establishing emissions inventories and clean air plans.

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*This paper has not been reviewed by the U.S. Environmental Protection Agency and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred.

SURVEY OF CLEAN AIR PLANS AND EMISSIONS INVENTORIES IN THE FEDERAL REPUBLIC OF GERMANY

Six of the 11 regions in the Federal Republic of Germany (FRG) have formally declared 21 areas in Germany to be HPAs (see Table 1 and Figure 1), and emissions inventories have been completed or are being prepared in 28 HPAs. Data on 24 of these areas were available for this report from publications and other sources. The total size of the 24 areas is about 9,500 km^2, and the total population is approximately 17.5 million (see Table 2). About 28% of the German population lives in these 24 areas, which amounts to only 3.8% of the total land mass of the FRG. The average population density in the HPAs is about 2,500 inhabitants, which is 10 times higher than the average population density for the entire FRG.

Clean air plans for HPAs have already been established in 3 of the 11 German regions. Several of the other regions are presently establishing clean air plans (e.g., Baden-Wuerttemberg, Bavaria, Hamburg, and Niedersachsen).

Clean air plans have already been completed in 9 of the aforementioned 28 HPAs. Comprehensive parts of clean air plans have been completed in more than half of these areas. Table 3 summarizes those areas where emissions inventories or clean air plans are being researched or have been completed.

TABLE 1. HPAS IDENTIFIED IN THE FRG

Region	Area (km²)	Inhabitants (thousands)	Population Density (Inhab./km²)	HPAs Declared	HPAs Number
Bayern	70,500	10,800	150	yes	8
Niedersachsen	47,400	7,300	150	no	(4)[a]
Baden-Wuerttemberg	35,800	9,200	250	no	(2)
Nordrhein-Westfalen	34,100	17,200	500	yes	5
Hessen	21,100	5,600	260	yes	4
Rheinland-Pfalz	19,800	3,700	190	yes	2
Schleswig-Holstein	15,700	2,600	160	no	-
Saarland	2,600	1,100	430	yes	1
Hamburg	750	1,700	2,300	no	(1)
Berlin	480	2,000	4,200	yes	1
Bremen	400	700	1.800	no	-
FRG	248,630	61,900	250		21 (7)

[a] HPAs in parentheses not yet formally declared as such.

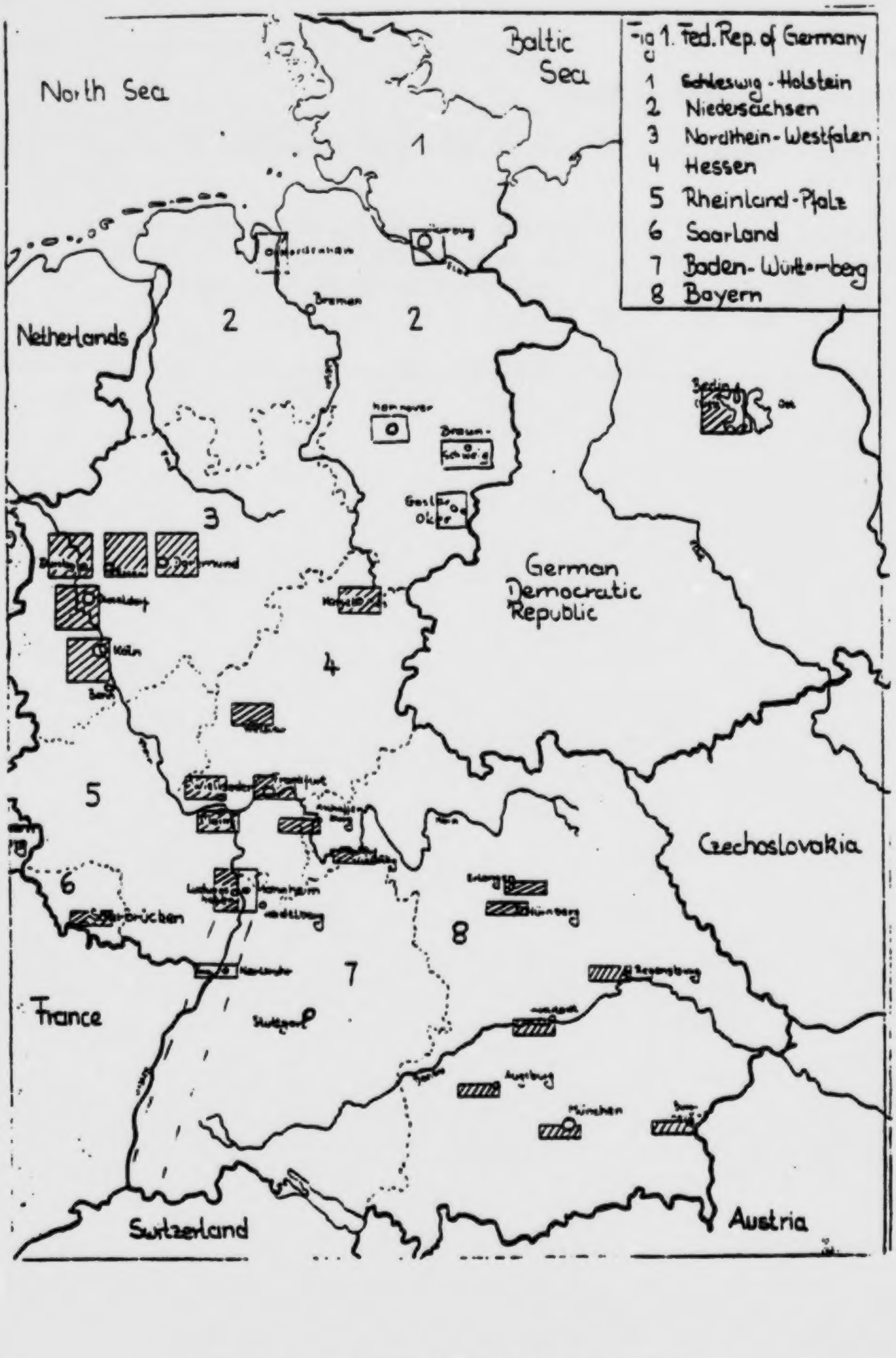

Fig 1. Fed. Rep. of Germany
1 Schleswig-Holstein
2 Niedersachsen
3 Nordrhein-Westfalen
4 Hessen
5 Rheinland-Pfalz
6 Saarland
7 Baden-Württemberg
8 Bayern
North Sea
Baltic Sea
Netherlands
German Democratic Republic
Czechoslovakia
France
Switzerland
Austria
Bremen
Hannover
Braunschweig
Goslar Oker
Dortmund
Düsseldorf
Köln
Bonn
Frankfurt
Mannheim
Saarbrücken
Karlsruhe
Stuttgart
Erlangen
Nürnberg
Regensburg
Augsburg
München
1
2
2
3
4
5
6
7
8

Figure 1. Federal Republic of Germany.

TABLE 2. SUMMARY OF EMISSIONS INVENTORIES IN THE FRG

Status	Number
Areas formally declared as HPAs	21
Areas in which emissions inventories have been completed or are in preparation	28
Areas in which emissions inventories have been completed or are in preparation, which have a total area of approximately 9,500 km^2 (approx. 3.8% of FRG) and a total population of 17.5 million inhabitants (approx. 28% of FRG)	24

TABLE 3. SURVEY OF HPAs

				Date of Completion	
Region	Area (km^2)	Inhabitants (thousands)	Population (Inhab./km^2)	Emissions Inventory	Clean Air Plan
Bayern (total)	2940[a]	3315	1100		
Aschaffenburg	740[a]	210	290		
Augsburg	230[a]	300	1300		
Burghausen	65[a]	25	380		
Erlangen-Nuremberg	431	770	1800	1982	
Ingolstadt	540[a]	170	320		
Muenchen	820[a]	1600	1900		
Regensburg	55	130	2400		
Wuerzburg	60[a]	110	1800		
Berlin (total)	480	1900	4000	1981[b]	
Hessen (total)	785	1600	2000		
Frankfurt+	466	1000	2100	1982	1983
Kassel	148	260	1800	1981	
Wetzlar	50	47	900	1980	1982
Wiesbaden+	120	290	2400	1978	1981
Nordrhein-Westfalen (total)	3200	6700	2100		
Dortmund+	712	1200	1700	1978	1978
Duisburg+	711	1300	1800	1975	1977
Duesseldorf	356	800	2200	1981	1982
Essen+	756	2000	2600	1980	1980
Koeln+	649	1400	2200	1972	1976
Rheinland-Pfalz (total)	205	400	1900		
Ludwigshafen	111	210	1900	1977	1980
Mainz+	94	190	2000	1978	1982
Saarland Sarbrueckken (total)	235	370	1600	1978/79[c]	
Baden-Wuertemberg (total)	319	590	1800		
Mannheim+	145	310	2100		
Karlsruhe+	174	280	1600	1980	
Hamburg	~750	~1700	~2300		
Niedersachsen	~500	~850			
Braunschweig					
Goslar/Oker				1981	
Hannover					
Nordenham					

[a] Estimate.

[b] Traffic source category.

[c] Excludes industrial source category.

CLEAN AIR PLANS

A clean air plan consists of the following elements: an emissions inventory, an air pollution (impact) inventory, an effects inventory, an analysis of causes, and a catalog of measures.

The emissions inventory is a detailed list of air pollutant emissions, indicating type, amount, spatial and temporal distribution, and emission conditions (source dimensions and location, waste gas volume flow, and temperature).

The air pollution (impact) inventory includes the results of measuring the most frequently occurring air pollutants present on a large (spatial) scale, i.e., SO_2, NO_x (NO and NO_2), CO, gaseous organic compounds, and dust (concentration and dust fall). These data are supplemented by air pollution impact data, which is determined from emissions inventory data via dispersion model calculations.

The effects inventory lists effects found in the area under investigation that are possibly precipitated air pollutants (e.g., forest damages, material damages, epidemiological effects).

The analysis of causes demonstrates the relationship between effects, air pollution impact, and emissions. Its aim is to determine measures to reduce air pollution emissions.

The catalog of measures results from the establishment of a clean air plan and outlines those measures that are most effective in maintaining air quality and reducing air pollution and its impact.

Emissions Inventories

Establishing the emissions inventory is a major element of a clean air plan. In the inventory, emission data are given for three source categories: (1) industrial installations, (2) domestic heating, trade, and small industries, and (3) traffic (mainly road traffic).

Industrial installations include all installations subject to licensing, such as power plants, chemical and petrochemical plants, steel works, nonferrous metal industries, and coke ovens. The second source category, referred to as "household," contains domestic heating facilities, fuel stations, dry cleaning installations, and printing offices. The traffic source category includes road, rail, water, and air traffic.

Due to the diversity of the industrial processes and sources, and the high number of air pollutants resulting from industrial plants, emissions data for the industrial installation source category must be investigated individually. This can be done either by having independent experts collect the necessary data pertaining to the different plant parts or by having the plant operator submit emission reports.

The emissions data for domestic heating, trade, and small industry installations are evaluated during investigations on the type, size, and spatial distribution of these installations. Emissions data are determined and correlated with local distribution by using emission factors for corresponding plant standards or standard plants. Because there are functional dependences between the technologies used and the emissions procedures followed, this type of assessment is sufficient.

The same approach applies to traffic sources. It is sufficient to determine the traffic population and the corresponding driving modes. Traffic emissions are then determined via emission factors for the different driving modes and related to local distribution and frequency of occurrence.

For the traffic and the domestic heating, trade, and small industry (household) categories, the emissions data are combined for line and area sources. Emissions data for the industrial category, however, are described mostly via point sources. The smaller emissions and the more uniformly distributed emissions of the industry are also described as line and area sources or even as volume sources. In emissions inventories or clean air plans, emissions are evaluated in terms of a grid that is 1 km x 1 km.

When an emissions inventory is established, there is no a priori limitation on the air pollutants included. Although the number of air pollution components for the traffic and the household categories are limited, the industry category includes many substances that can lead to air pollution impacts. The number of these substances in the industry category may be 1,000 or more. Table 4 gives a rough summary of the components considered when analyzing the different source categories.

Emissions inventories in the FRG formerly consisted of investigations conducted by independent experts of all plants, sources, substances, and emissions. This practice has now changed; the plant operators must now declare all emissions data annually. Thus, an actual updating is guaranteed every year.

Updating emissions data for the traffic and the household source categories is performed 5 yr by the local administration for the respective area. Clean air plans have to be established or reviewed every 5 yr, too, by the regional administration and its institutions.

The approach for investigating an emissions inventory in The Netherlands and Belgium is very similar to that practiced in the FRG.

Status of Investigation

Presently, clean air plans with emissions inventories have been published for the following regions in the FRG: Cologne, Duisburg, Dortmund, Ludwigshafen, Essen, Wiesbaden, Duesseldorf, Mainz, and Wetzler. In addition, emissions inventories are available for the following areas: Berlin (traffic only), Saarland (traffic and domestic heating only), and the Mannheim, Karlsruhe, Goslar/Oker, Nuremburg, and Frankfurt regions. Emissions inventories are being prepared for Hamburg and Berlin (industrial emissions), and for the

TABLE 4. AIR POLLUTANTS IN EMISSIONS INVENTORIES INVESTIGATED

	Source Catategory			
Pollutant	Industry	Small Industry	Domestic Heating	Traffic
SO_2	X	X	X	X
NO_x	X	X	X	X
CO	X	X	X	X
Fluorine compounds	X	X	X	
Chlorine compounds	X	X	X	
Total dust/particulates	X	X	X	
Lead/lead compounds	X			X
Soot				X
Organic compounds	X	X	X	X
Aldehydes	X		X	
Benzopyrene	X			
All other organic compounds	X			
All other inorganic compounds	X			
All other types of dusts and particulates	X			

Kassel region. Clean air plans are being prepared for the Kassel and Frankfurt regions and may be available towards the end of 1983.

The first updated clean air plan in the FRG will be available in 1983 for the Cologne region. It will be particularly interesting to compare the results of the (experts) investigations for the first clean air plan with those of the emissions declarations, which will be included in the second clean air plan for the region.

Emissions inventories for areas outside the FRG are available for Gent and Liege, Belgium, for five areas of The Netherlands, and for Denmark. This listing may be incomplete as requests for emissions inventories were only made recently to several nations.

Results with Respect to Oxidants and Their Precursors

Selected results of one emissions inventory, a general survey of data available in the Cologne clean air plan, are given in Tables 5 through 8 and Figures 2 through 4. The survey includes the topics of this workshop.

Table 5 presents data from the major emissions groups, particularly organic compounds, for the three source categories. Table 6 presents the major inorganic compounds, particularly NO_x, for the three source categories. Table 7 presents the organic compounds, subdivided into HCs and halogenated HCs, for the industry source category. Table 8 is a rough survey of halogenated HCs.

TABLE 5. EMISSIONS DATA REPORTED IN THE COLOGNE CLEAN AIR PLAN

Source Category	Emissions (thousand tons/yr) Inorganic Compounds	Organic Compounds	Dust and Particulates	Total for Category
Industry	298	84	25	407
Household	107	6	5	118
Traffic	134	5	0.5	140
Total for all categories	539	95	30.5	665

TABLE 6. INORGANIC COMPOUNDS REPORTED IN THE COLOGNE CLEAN AIR PLAN

Source Category	Emissions (thousand tons/yr) NO_x	SO_x	CO	Other Species	Total for Category
Industry	76	147	69	6	298
Household	4	18	84	1	107
Traffic	10	1.5	123	0	134
Total for all categories	90	166.5	276	7	539

TABLE 7. ORGANIC COMPOUNDS FROM INDUSTRIAL SOURCES AS REPORTED IN THE COLOGNE CLEAN AIR PLAN

Compound	Emissions (tons/yr)	(% of Total)
Hydrocarbons	56,127	67
C_1-C_4-HC	29,780	35
Fuel-HC (200JC)	10,771	13
Aromaties	9,414	11
Fuel-HC (200JC)	1,970	2
Other compounds	27,990	33
Halogenated HC	5,725	7
Other	22,265	26
Total of all organic compounds	84,177	100

TABLE 8. HALOGENATED HCs INDUSTRIAL SOURCES AS REPORTED IN THE COLOGNE CLEAN AIR PLAN

Compound	Emissions (tons/yr)	(% of Total)
Vinyl chloride	1,335	23
Dichloromethane	1,209	21
1,2-Dichloroethane	1,161	20
Trichloroethylene	504	9
Ethyl chloride	167	3
Carbon tetrachloride	167	3
Perchloroethylene	152	3
Other	1,030	18
Total of all HCs	5,725	100

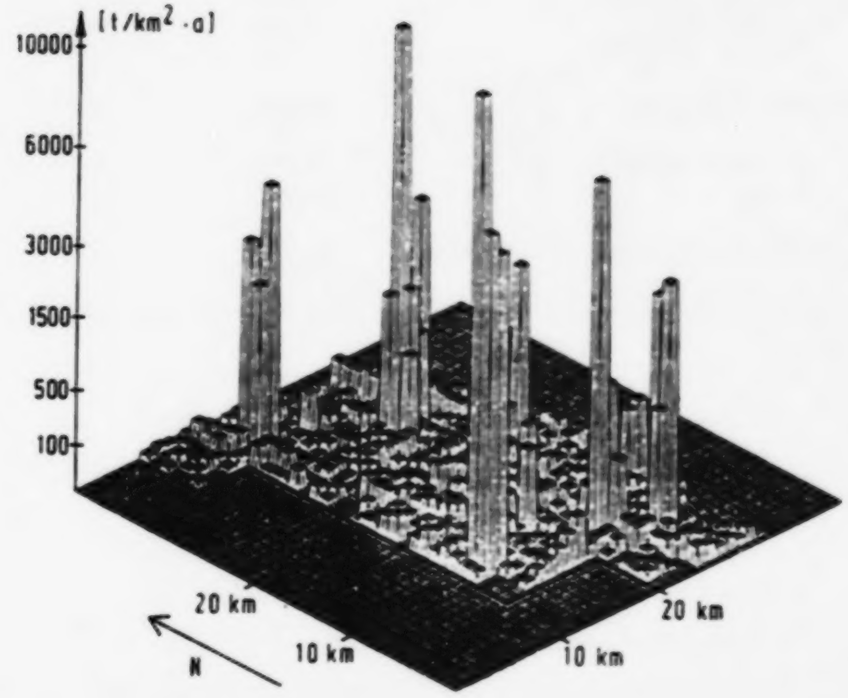

Figure 2. Total annual emission of nitrogen oxides.

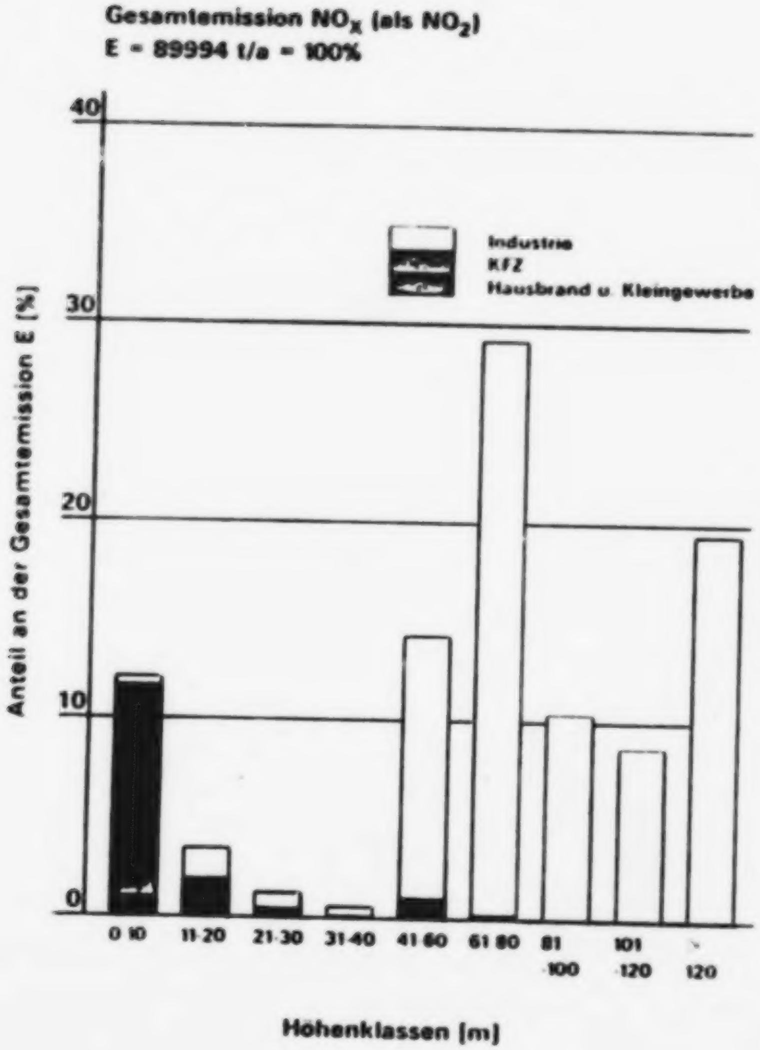

Figure 3. Total emissions of NO_x (as NO_2).

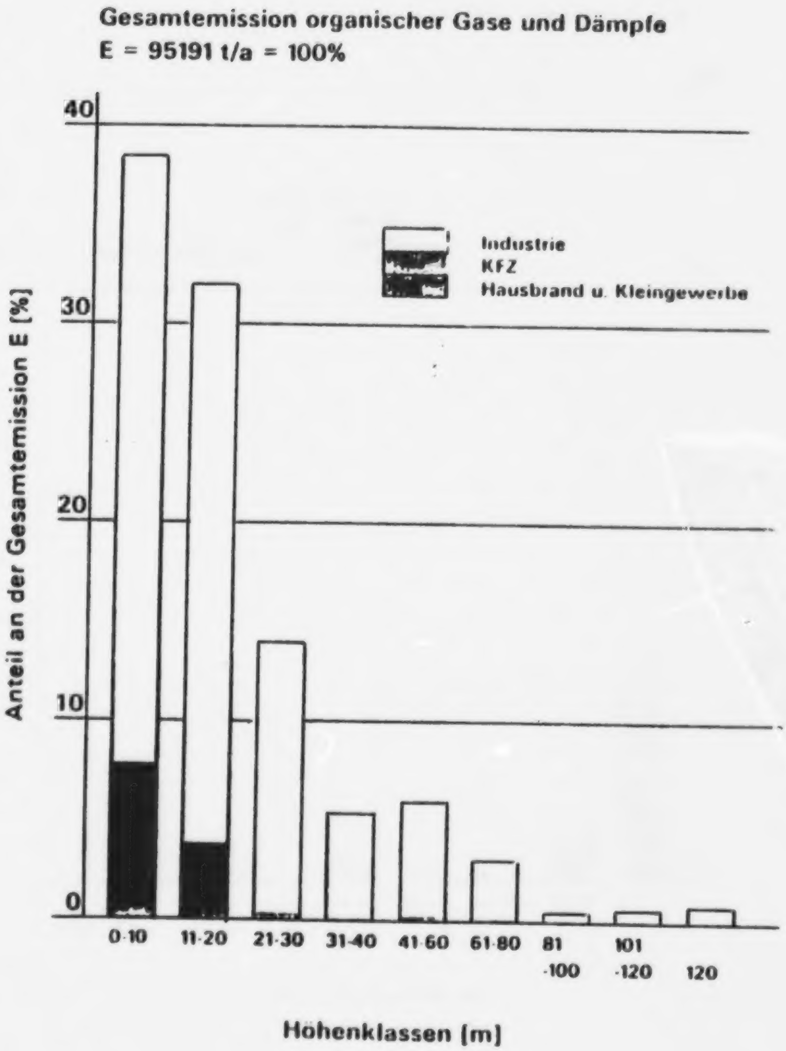

Figure 4. Total emissions of organic compounds.

Figure 2 is a three-dimensional display of the NO_x (as NO_2) distribution. The grid size is 1 km x 1 km, and the peak heights represent the total emissions per grid. Figures 3 and 4 are displays of NO_x (as NO_2) and organic compound emissions, respectively, for different height categories. Although most NO_x are emitted at heights over 50 m, most organic compounds are emitted at heights below 50 m.

The most important data with respect to this workshop are emissions data for NO_x, HCs, and halogenated HCs in the FRG and Europe. Tables 9 through 12 survey emissions in FRG for the following source categories: industry, household, and traffic. Emissions from sources over 100 m high were excluded where possible.

Table 9a shows a total NO_2 emission of about 675,000 tons per year for all areas with established emissions inventories. About one-third (200,000 tons per year) of these emissions occur at heights over 100 m. In comparison to the total NO_x emissions from these HPAs, the total NO_2 emissions for the FRG is 4 to 5 times higher (3 million tons of NO_2 per year). This is due to the large amounts of emissions from traffic and domestic heating sources, as well as emissions from large power stations outside the HPAs. Estimates of NO_x emissions over large stacks (i.e., with heights greater than 100 m) are 5 to 6 times higher than those for emissions within HPAs.

Table 10a shows the corresponding data for SO_2. Because SO_2 is less important in oxidant formation than NO_x, these data are given for comparison only. The total SO_2 emissions from HPAs is approximately 1.2 million tons per year, one-third of the total SO_2 emissions for the FRG. More than two-thirds of the emissions occur via stacks over 100 m high. The emissions data for organic compounds are given in Table 11a. About 480,000 tons of organic compounds per year are emitted in the HPAs, only one-third to one-fourth of the total emissions for the FRG. Only a very small part (less than 1%) is emitted via stacks over 100 m high.

Industrial HC emissions amount to 270,000 tons per year, or 60% of the total emissions for all organic compounds (see Table 12). Of the HCs emitted by industry, only 8% are halogenated HCs. These figures refer solely to emissions data for HPAs. Corresponding figures for the FRG are not available. Although the HCs emitted at higher altitudes cannot be extracted from published data, these values can be extracted from the base data used to establish emission figures.

Some corresponding data for NO_x, SO_2, and organic compound emissions in other nations are presented in Tables 9b through 11b.

TABLE 9a. NO_2 EMISSIONS IN THE FRG

Region	HPA	Emssions (thousand tons/yr) Industry	Household	Traffic	Total for HPA	Emissions at >100 m
Bayern	Nuremberg	14.6	1.9	6.6	23.1	10
Berlin	Berlin	+[a]	+	4.0	+	+
Hessen	Frankfurt	(37)[b]	(4)	13.0	(54)	+
	Wetzlar	0.4	0.2	0.6	1.2	-
	Wiesbaden	3.2	0.9	2.3	6.4	(1)
Nordrhein-Westfalen	Dortmund	81.1	2.7	11.8	95.6	45
	Duisburg	75.1	3.3	12.7	91.1	35
	Duesseldorf	24.2	1.9	8.6	34.7	8
	Essen	137.0	3.8	16.6	157.4	60
	Cologne	75.8	4.5	9.7	90.0	24
Rheinland Pfalz	Ludwigshafen	23.5	0.6	2.9	27.0	(14)
	Mainz	9.1	0.6	2.1	11.8	1
Saarland	Saarbrueckken	+	1.3	3.2	(10)	+
Baden-Wuerttemberg	Karlsruhe	11.1	0.5	1.7	13.3	7
	Mannheim	55.1	0.4	1.1	56.6	37
Total for all HPAs		550	27	98	675	(200)
Total for FRG, 1978		1,520	140	1,340	3,000	(1,100)

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[a] Data not available indicated by "+".
[b] Numbers in parentheses are estimates.

TABLE 9b. NO_2 EMISSIONS IN OTHER EUROPEAN COUNTRIES

HPA	Emissions (thousand tons/yr) Industry	Household	Traffic	Total for HPA	Estimates at >100 m
Belgium					
Gent	20.4	1.0	5.3	26.7	(10)[a]
Liege	13.8	0.8	3.5	18.0	4
The Netherlands					
South Holland				105.3	
North Holland				62.0	
Utrecht				45.6	
Oberijssel				20.0	
Denmark	96	66	64	226	
West Germany	1,520	140	1,340	3,000	(1,100)

[a] Numbers in parentheses are estimates.

TABLE 10a. SO_2 EMISSIONS IN THE FRG

Region	HPA	Emissions (thousand tons/yr) Industry	Household	Traffic	Total for HPA	Emissions at >100 m
Bayern	Nuremberg	20.8	5.6	0.4	26.7	20
Berlin	Berlin	+[a]	+	0.3	+	+
Hessen	Frankfurt	(75)[b]	(10)	0.7	(86)	+
	Wetzlar	0.7	0.4	0.1	1.2	-
	Wiesbaden	5.9	2.7	0.2	8.8	(1)
Nordrhein-Westfalen	Dortmund	112.3	12.7	1.3	126.3	80
	Duisburg	193.4	15.1	1.9	210.4	100
	Duesseldorf	29.1	5.0	0.5	34.6	8
	Essen	147.5	17.6	1.3	311.9	230
	Cologne	75.8	18.5	1.5	167.5	45
Rheinland-Pfalz	Ludwigshafen	32.8	1.4	0.2	34.4	12
	Mainz	13.1	1.2	0.1	14.4	1
Saarland	Saarbrueckken	+	4.7	0.4	(10)	+
Baden-Wuerttemberg	Karlsruhe	60.3	1.9	0.4	62.6	44
	Mannheim	63.9	1.5	0.2	65.6	42
Total for all HPAs		1,050	100	10	1,160	(600)
Total for FRG, 1978		3,000	450	100	3,550	(2,500)

[a] Data not available indicated by "+".
[b] Numbers in parentheses are estimates.

TABLE 10b. SO_2 EMISSIONS IN OTHER EUROPEAN COUNTRIES

	Emissions (thousand tons/yr)			Total for HPA	Emissions at >100 m
HPA	Industry	Household	Traffic		
Belgium					
Gent	130.2	5.7	0.7	136.6	65
Liege	41.3	3.6	0.3	45.2	25
Total for Belgium	690	103	16	808	u.a.
Denmark	295	172	5	472	
West Germany	3,000	450	100	3,550	(2,500)

TABLE 11a. ORGANIC COMPOUNDS IN THE FRG

Region	HPA	Emissions (thousand tons/yr) Industry	Household	Traffic	Total for HPA	Emissions at >100 m
Bayern	Nuremberg	6.7	1.1	5.2	13.0	1
Berlin	Berlin	+[a]	+	4.5	+	+
Hessen	Frankfurt	(18)[b]	(22)	9.1	(49)	+
	Wetzlar	0.5	0.1	0.5	1.1	-
	Wiesbaden	9.4	1.0	2.1	12.5	(0.9)
Nordrhein-Westfalen	Dortmund	4.4	4.9	10.2	19.5	(0.2)
	Duisburg	150.7	12.4	9.1	172.2	0.5
	Duesseldorf	2.7	3.8	6.4	12.9	0.1
	Essen	29.3	9.4	13.7	52.4	0.1
	Cologne	84.1	6.3	4.8	95.2	1.4
Rheinland-Pfalz	Ludwigshafen	12.5	1.0	2.1	15.6	0.1
	Mainz	2.4	0.5	1.4	4.3	-
Saarland	Saarbrueckken	+	2.1	3.4	(6)	+
Baden-Wuerttemberg	Karlsruhe	9.4	0.3	2.3	12.0	0.1
	Mannheim	6.9	0.3	1.9	9.1	0.1
Sum HPAs		340	65	77	480	4
FRG (1978)		480	630	650	1,750	(13)

[a] Data not available indicated by "+".

[b] Numbers in parentheses are estimates.

TABLE 11b. ORGANIC COMPOUNDS IN OTHER EUROPEAN COUNTRIES

HPA	Emissions (thousand tons/yr) Industry	Household	Traffic	Total for HPA	Emissions at >100 m
Belgium					
Gent	6.2	0.3	1.9	8.4	(1)[a]
Liege	5.3	0.1	1.6	7.0	(1)
Total for Belgium	12	0.4	4	16	2
The Netherlands					
South Holland				130	
North Holland				88.8	
Utrecht				38.7	
Gelderland				87.3	
Oberijssel				64.0	
Total for The Netherlands				409	
West Germany	480	630	650	1,750	(13)

[a] Numbers in parentheses are estimates.

TABLE 12. HYDROCARBONS AND HALOGENATED HYDROCARBON EMISSIONS[a] IN FRG (FIGURES IN BRACKETS ARE ESTIMATES)

		Industry			
Region	HPA	HCs	HCs at >100 m	Halogenated HCs	Halogenated HCs > 100 m
Bayern	Nuremberg	6.0	+[a]	3.1	+
Berlin	Berlin	++[b]	++	++	++
Hessen	Frankfurt	17.4	++	3.7	++
	Wetzlar	0.3	-	0.2	-
	Wiesbaden	3.0	+	1.7	+
Nordrhein-Westfalen	Dortmund	1.8	+	0.2	+
	Duisburg	145.0	+	1.5	+
	Duesseldorf	0.5	+	0.4	+
	Essen	19.3	+	2.0	+
	Cologne	56.1	+	5.7	+
Rheinland-Pfalz	Ludwigshafen	4.8	+	1.8	+
	Mainz	1.5	+	0.2	+
Saarland	Saarbrueckken	++	++	++	++
Baden-Wuerttemberg	Karlsruhe	8.3	0.1	0.2	-
	Mannheim	3.6	-	0.6	-
Total for all HPAs		270	?	33	?

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[a] Emission in thousand tons per year.
[b] Data not available are indicated by "+".
[c] Data not available from publication are indicated by "++".

SUMMARY

Detailed emissions inventories are already available or will shortly be available (within this or the next year) for about 3.8% of the FRG, which includes about 28% of all German inhabitants.

During detailed investigations of the emissions inventories of the three source categories--industrial; domestic heating, trade, and small industry, and traffic--emissions data for more than 1,000 substances were collected.

Emissions inventories that have been published and are already available for 15 regions indicate that:

- More than 680,000 tons of NO_x are emitted per year (81% from industry, 4% from domestic heating, and 15% from traffic sources); this is about 22% of the total NO_x emissions per year for the FRG.
- Nearly 0.5 million tons of organic substances are emitted per year (70% from industry, 14% from households, and 16% from traffic sources); this is 27% of the total organic compound emissions for the FRG.
- About 270,000 tons of HCs and about 22,000 tons of halogenated HCs are emitted per year due to industrial emissions.

Although 37% of the nitrogen compounds are emitted at heights of over 100 m, less than 1% of the organic compounds are emitted at such heights.

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APPENDIX A. GUIDELINES FOR EMISSION INVENTORY PRESENTATIONS

Data base name/source	Various emissions inventories in FRG.
Reasons for inventory development?	Air quality management, clean air plans.
Who collects the raw data? (private industry, national/provincial government, etc.)	Independent experts by order of lander administrations (provincial) or plant operators.
How is raw data collected? (questionnaire, permit system, inspection, other)	Individual inspection and/or emissions declaration, licensing procedures, emission factors.
How frequent are data updated?	Industry, yearly; all others; every 5 yr.
Are updates legally required?	Yes, legal regulations ( )
List legal or confidentiality restrictions which may prevent release of the data	Restricted for use by local authorities or lander administrations only.
Area of coverage	Total approx. 10,000 km^2 (indiv. 50...50 km^2).
Coordinate system	Gauss-Kruger grid (rectangular grid)
Point source information; define a point source	Single source with great emission, e.g., large stacks
A) Raw data collected:	
List stack information	height, diameters, flow rate, temperature,
List major contributing source categories (industries)	chemical, petrochemical, metal industry
List types of raw data collected and temporal resolution where appropriate	, SO_2-emissions from fuel consumption and sulfur content; 1 h.
Spatial resolution	1 m, if necessary.
Dates of available data	Previous year.
B) Emission estimates:	
List pollutant species	SO_2, NO, NO_2, CO, HC, Pb, and some 1,000 others.
Temporal resolution of calculated emissions	One hour.
List information available for temporal apportionment	Daily distribution of traffic, operating rate of day, week, year.
List percentage of emissions estimated by following methods:	Very rough estimates.
____ Standard emission factors with specific plant information	20%
____ Nonstandard emission factors with specific plant information	30%
____ Source test	20%
____ Material balance	20%
____ Other, specify	10%
What emission factors, if any, are used?	Traffic, domestic heating, small industry (corresponding to standard plants).
List publication describing emission factor development program.	

Are reported emissions controlled or uncontrolled?	Partly controlled.
Are control equipment and efficiency information available?	For individual point sources, only.
Describe method of estimating volatile organic compound emissions	APTI--formula, e.g., tanks breathing.
Area source information; define an area source	For example, 1,000 m x 1,000 m (domestic heating, traffic if not as main street described)
A) Raw data collected	
List major contributing source categories	Traffic, domestic heating, small industry, leakages of chemical/petrochemical plants.
List subclasses of stationary area mobile sources	Motorcycles, trucks, vessels, aircraft.
List types of raw data collected, spatial and temporal resolution where appropriate	Type, size, and spatial distribution of domestic heating, fuel sales, local distribution, and frequency of traffic; 1 km, 1 h if necessary.
Dates of available data	1970-1982, depending on area of investigation.
B) Emission estimates:	
List pollutant species	SO_2, CO, NO, NO_2, HC, particulates, others
Temporal resolution of calculated emissions	1 h if required.
List information available for temporal apportionment	Annual, seasonal, weekly, daily, hourly distributions.
Describe grid system or spatial resolution	Longitude/latitude in meters.
List information available for spatial apportionment	Housing location, street.
Are published standard procedures used for temporal and spatial allocation and emission calculations?	Publication 280 of ?
If yes, list major references	Regulation ?
Describe method of estimating volatile organic compound emissions	API

General

Comment on completeness	The completeness of SO_2 is better than of NO and NO_2 (mostly NO_x as NO_2). Point sources better than area sources.
Comment on currentness	Point source data are mostly 1 yr old. Area source up to 5 yr old.
Summarize quality assurance program	According to updating procedure (emissions declarations). Control and comparison of data by lander administration.
Who is responsible for data quality?	German lander administration.
Are source inventory data handled manually or by computer?	All data handled on computers.
Attach detailed record formats	

APPENDIX B. GUIDELINES FOR AEROMETRIC DATA PRESENTATIONS

Surface Air Quality

Data base name/source	
Area of coverage	
Total number of monitoring sites	
Spatial distribution (attach site map)	
Year of record	
Check available site information:	
____ physical location (lat-long, UTM)	
____ geographic location (state/province/	
____ elevation (MSL, AG)	
____ classification (i.e., urban, rural suburban, remote)	
____ environment of site	
____ descriptive information	
____ dominating influence (i.e., industrial, residential, mobile)	
____ other, specify:	
Parameters (attach table of measured parameters, associated equipment type/analysis method and temporal resolution)	

Upper Air Quality

Data base name/source	
Spatial distribution (attach standard of flight paths)	
Year/date of record	
Parameters (attach table of measured parameters, associated equipment type/analysis method and temporal resolution)	
Spatial resolution	

Surface Meteorology

Data base name/source	NOAA
Area of coverage	20° - 55°N, 65° - 130°W
Total number of stations	-
Spatial distribution (attach site map)	See map.
Site information available	-
Parameters measured	Standard airways observation
Time interval of measurements	Hourly
Year/date of record	Aug. 1979: June, July, Aug. 1980

Upper Air Meteorology

Data base name/source	NOAA
Area of coverage	20° - 55°N, 65° - 130°W
Total number and type of stations	
Spatial distribution (attach site map)	See attached map
Site information available	
Year/date of record	Aug. 1979; June, Julv, Aug. 1980
	0000, 12; 0600, some; 1800, special
Rawinsonde release	
parameters measured	Standard RAOBs
frequency of release	12 hours - some 6 hours
maximum level	500 mbar
Pibal release	
parameters measured	
frequency of release	
maximum level	
Others, specify:	

SESSION IV

MODEL EVALUATION

April 13, 1983

Session IV consisted of panel discussions on six selected models. These presentations are summarized in Session V.

SESSION V

MODEL EVALUATION
INDIVIDUAL DISCUSSION GROUPS

April 14, 1983

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STEM MODEL

P. Builtjes

Yesterday, we discussed the STEM model by Gregory Carmichael. Very briefly, it is an Eulerian grid model used to calculate oxidants over an area of, say, 1,000 x 1,000 km^2, with the emphasis on vertical resolution. The model has about 10 or 11 layers in the vertical direction.

As I understand it, the model has four objectives. In order of importance, the main objective is that the model be used to provide a detailed depiction of field studies. That is the reason the model has about 10 or 11 layers, so that you can clearly see and follow plumes, their mixing, and their interaction with the surroundings.

Another objective is that the model be used to determine the influence of regional scales on global scales. That is one reason why the model has a larger height than the other models, a height of about 8 km, with the troposphere more or less.

The third objective is to use the model to investigate specific processes. So, you can use it as a kind of learning model, with the purpose of incorporating processes if they appear to be of importance in a simpler model.

Finally, you can also use the model for control strategy purposes. However, I think the main objectives are the three I have just mentioned.

The STEM model requires a considerably larger computer memory than the Eulerian SAI model or Lamb model.

Different modules of the model have been tested, but that does not mean that the total model is fully operational. It has been estimated that it will take 3 yr before the model can be made fully operational, that is, if the funding is available to do so.

A major problem with this kind of model, as with the Lamb model, is the incorporation of clouds, that is, the real dynamic direction inside the model itself with clouds.

The difference between the Carmichael model and the Lamb model is that the Carmichael model is fully Eulerian. The Lamb model has a special treatment of interchange between vertical layers, but you can say it is not fully Eulerian.

In our panel meeting, we also concluded that there will be a problem in the way in which these models have to be validated, especially due to the large grid size used in this kind of Eulerian model.

We also discussed whether a region of about a 1,000 x 1,000 km^2 exists in Europe with sufficient input data, say emissions and aerometric data, to validate the models. Although there was not complete agreement about whether such an area exists, my opinion is that it could be done. There are difficulties, but data are available.

Whether it is necessary to incorporate certain phenomena into the Carmichael model and to what level of sophistication the phenomena can be incorporated cannot yet be decided, because no real sensitivity runs and no validations runs have been made. So, we do not know exactly how sophisticated the treatment of clouds should be, whether it is necessary, and whether it has any influence in a real validation approach.

Let's summarize by considering the objectives of the workshop.

The first item concerns the state of the art of existing emissions inventories and the plan to complete these inventories as model input data. Emissions inventories already exist for the corridor units in the United States, which are complete between (marks), and they are available in principle for parts of Europe, also between (marks).

We did not discuss the second item, regarding the refinement of the best available models.

The third item involved the merits of selected models as the bases for developing control strategies. As I said, the Carmichael model can be used for control strategies, but the major objectives are different.

The fourth item, the chemistry, was not discussed, and there is no recommendation for the chemistry. There are several chemical mechanisms. Of course, the model is modular. You can take chemistry out or leave it in if you like.

The fifth point was to examine whether the aerometric data base, including boundary condition data, is sufficient and, if not, to formulate a plan to obtain the missing data. I do not know about the United States, but in the European situation we mainly lack HC measurements, field measurements of HCs, especially somewhere along the boundaries of your areas.

The sixth point was to determine when the model will be fully operational. As I said, about 3 yr from now, 1986.

A. Galli: Is that when it will be a validated model or just available for use?

P. Builtjes: It will be a validated model around then.

B. Dimitriades: I thought the question was whether the model would be operational.

P. Builtjes: It was stated as fully operational, and we interpreted the term "fully operational" to mean whether we could use the model to develop control strategies and whether it could be validated.

The final point involved defining the modeling domain in terms of the available emissions and aerometric data, the topography, and the model's capability. As far as we are concerned, the domain should be the northwestern part of Europe, because most of the available input data are for that area.

DISCUSSION

E. Runca: Regarding the validation of the model, it is not clear to me what data are necessary. Could you comment on that?

P. Builtjes: Perhaps Gregory Carmichael should comment.

G. Carmichael: Although we tried to discuss that at the end of the session yesterday, we really did not come up with a concrete assessment of a validation plan for such models. We have talked about the validation procedure, but I do not have a detailed plan for that procedure.

We mentioned some of the difficulties in validating large-scale models, implying that, with a plan, you have look at data requirements and whether there are enough observational data to really verify field study data, and we pointed out deficiencies in the HC measurements as one area of concern. In terms of a detailed plan, that is included in the 3-yr period for the detailed verification plan. Until we have a better idea of how to verify large-scale regional models, we need that plan first and then an assessment of--now with an understanding of your data bases, then we can make a better assessment of that. I think it would be premature for me to speculate.

A. Galli: What do you need to make your model operational? Do you need meteorological data, chemistry data?

G. Carmichael: Do you mean to exercise the model?

A. Galli: Right, what is implicit in the model? What do you already have there, and what is needed?

G. Carmichael: If you are really doing an event assessment and it is a three-dimensional model, you need good vertical resolution. So, the best test would be under conditions where you have as much upper air data as possible, so that you can specify the transport as well as possible. That would mean conditions where you not only have as high a density of upper air data as possible but also a frequency of upper air data at least four time a day for such, and you may only have data for twice a day.

So, some of the deficiencies are that. If you select a certain time period, there may have been field studies that have been conducted that there are data for that time period.

A. Galli: Do you need anything else besides upper air data?

G. Carmichael: You need land use data, but you can presumably get that, and surface roughness and emissions data. If you select the proper area, there is apparently adequate emissions data for HCs and NO_x.

A. Galli: So the key is the upper air?

G. Carmichael: Well, that is one. You also want to specify the transport as well as possible. This model takes the raw data through a preprocessing. Whether it is done by our preprocesses or other people's preprocesses, you end up with a description of the met field.

If you are really operating the model and looking at it from the standpoint of whether you want to look at the effects of clouds, etc., there is a real question of how you get those data, how you locate the clouds in the region, and whether those data are available for the Europeans. This is an area where we have ideas and other people have other ideas, but there is no good way of doing it or no implemented way of doing it.

E. Runca: Where would you put the emphasis for your concentration--HC measurements, oxidant measurements to distinguish between O_3 and NO_2?

G. Carmichael: In this type of model where you are looking at many different species, you would obviously want O_3 measurements and all the species that significantly influence O_3 measurements. You would want NO, NO_2, and HC measurements as much as possible. It depends on what level. If you do a total validation, you obviously want as much data and related species data as possible, so that you can tell whether you are predicting O_3 correctly, how you are predicting the other species related to O_3, or whether you are predicting all of the relationships in a similar biased fashion.

D. Jost: Are the HC measurements going to be used for testing the output of the model or do you need other input in addition to the emissions?

G. Carmichael: You want it from many standpoints. First, it gives you a better idea of boundary conditions. Wherever you put this region, you need some idea of boundary conditions. Since you are predicting HCs, you also want to test the model on how it does the HCs, their influence, some of the photochemistry.

G. Whitten: Your report indicates that part of the validation procedures has already occurred in that you tested the numerical scheme for solving the chemistry package. In my judgment, the numerical scheme does not work very well. Is there some consideration of using in the next 3 yr alternate numerical schemes for the chemistry-solving package?

G. Carmichael: I do not agree with your statement. In what sense is the chemistry scheme calculation--what are you basing that observation on?

G. Whitten: The results presented in your paper indicate that, as you function the time, the present scheme diverges from the base case. In such a case, for application to a mesoscale over a period of several days, you want something that is more stable over long time periods and numerical algorithms that can be better performed. I was just wondering if that was of concern to you?

G. Carmichael: I still do not understand what you are talking about.

G. Whitten: You had a figure in your report that showed various time steps being used for the chemistry.

G. Carmichael: CSMP.

G. Whitten: What did you mean by CSMP?

G. Carmichael: We used a stiff integrator, in this case in the package.

G. Whitten: What is CSMP?

G. Carmichael: It is a simulation package. It's a stiff solver, so we'll take that to be an analytical--we'll take that to be the correct solution.

If I can clarify. If you compared the integration of the transport species of the chemical species over a 5-h period by using that technique, you would get the types of divergence, 10% or so after 5 h. However, our transport step involves 15-min intervals, and this chemistry is embedded in those transports only over 15-min intervals at 0.1 min. So, when you locate each individual time step from those initial conditions, you have very good agreement between those at that time scale, that resolution.

G. Whitten: My experience as a chemical modeler is that it is very important to follow a chemical scheme over a period of several days, and to integrate over 15-min intervals even over many days. And that diverged from the STEM solving package, I found it to be of considerable importance and of some concern.

There are other methods of solving the chemistry package that do not diverge from the stiff package.

D. Jost: Could we come to this question again in case it would be necesary for the further procedure after this meeting? If people feel that this question needs to be clarified now, then we could come back it. Would you both agree?

G. Carmichael: Yes.

G. Whitten: Yes.

B. Dimitriades: I just want to bring to the conferees' attention the possibility for confusion here. I want to go back to the comment about an operational model and full validation. It seems to me the point is when a model will be ready to be used by OECD in the subsequent evaluation program. By that time, the model should be fully operational, validated, verified, or what? Perhaps someone could define this for us. What is fully validated? What is fully operational? What is evaluated and what is verified? This may cause some confusion because I hear that the model should be fully validated in time to be used by OECD in another validation program.

D. Jost: Several people are asking for the floor. Perhaps I should attempt to answer this.

S. Reynolds: As part of any model application at this stage, epecially for regional models, any modeling study has to include an evaluation step before going into a control strategy analysis, and I think it is really incumbent upon the model users to demonstrate that the model is working adequately, whatever that means. So, it would be helpful if there was some previous experience. Nevertheless, it will be incumbent upon the user to provide evidence in the specific context, in this case of whatever the OECD application would be, that in Europe the model is operating properly. So that first and the application subsequent.

B. Dimitriades: You mean to do this with field data, I presume? A small-scale evaluation of the model?

S. Reynolds: Certainly some kind of evaluation. Ideally, some special measurement study might be mounted, given that existing data have not been collected, with the objective of evaluating or validating a model. It is possible to use the existing data to get an idea of how well the model might work, and that might be a first evaluation step, perhaps pointing the direction to further field studies that might provide a more comprehensive evaluation, perhaps a little further down the road in time.

B. Dimitriades: Perhaps Mr. Lieben could speak?

P. Lieben: Yes, I support this interpretation. What we are trying to do in OECD is to arrive at some later stage where we can recommend to member countries one model or several models that can be used for developing and implementing control strategies. That is the goal.

Having reviewed the situation during these 3 days, we now have to determine what needs to be done before we arrive at that point. The question is simple, but the answer is not so simple. That is what I would think is a goal in order to present to the member countries a model upon which they can rely. I would not say it would be absolutely perfect, but it would give sufficiently reliable answers needed for control strategies. What we have to do now is the work to be able to arrive at that point.

D. Jost: This includes some steps for validation and evaluation, and it depends on the state of the art of those models for which steps have already been made, whether or not they have already been validated or evaluated. As for the question, against which data should those evaluations be done, it is mostly atmospheric data, but this is not necessary. Other possibilities could be given against which such an evaluation could be done.

A. Christie: Are we perhaps oversimplifying this? In terms of the complexity of the models being discussed, you can tune the model to fit almost any data base that is available. So, it is fairly obvious that any kind of model development is going to involve each of these models. Each of these models is developed according to what will occur.

It is also fairly obvious that each of these models will have to have a separate evaluation and data base, which is probably a great deal finer than what will be available for evaluating the final total model. If you are going to put in more than one complex study in chemistry, you are probably going to need an evaluation set for the chemistry module, an evaluation set for the advection modules, and something that is going to evaluate whether you have cloud processes. Then, you can start looking at an evaluation data set.

If each person talks about evaluating a model in a different way with a system that tunes the model to what he/she is suggesting as an evaluation set, that really is not going to be a proper evaluation at all. I think the whole question is posed in much too general a way.

The development of a model of this kind is going to involve whole strategies of evaluation between the cost of putting all these separate modules together.

A. Galli: I look at this from a slightly different standpoint; that is, that Dr. Builtjes essentially said that the Carmichael model had not been validated, that essentially no sensitivity work has been run on the model, and that it would roughly be 3 yr before the thing would be completed for any real use for control strategies.

Regardless of what scheme is utilized to validate test sensitivity, the time frame that we are looking at in this whole control strategies program is 2 to 3 yr before the thing is set up, completed, and capable of giving someone something.

If the model is not going to be ready for 3 yr, we have obviously got a problem in the time frame that is being set up to begin with. As we look at each of the models, I think we are going to see some inherent problems, either from a technical standpoint of where they are in their development, not that their development is wrong or that their purpose is wrong, but that they are off in the time frame that we have set up, perhaps in OECD. As we go through some of these things, that will become very obvious to us, regardless of the technical plans that are being put forth. The bottom line is that where they are in the development is going to be a self-limiting factor as far as their use for the project, regardless of whether they are scientifically sound or unsound. It is a little hard to sit down, tear down, and build up a model when it is really not ready and it is not going to be ready in the time frame that you need it in.

D. Jost: That is right; I think this time frame is important. Nevertheless, although it is the object and OECD could resolve and propose some strategies, one needs to be aware of the development of these models that will be used in the OECD project and that they and will not be the final end in development in this field.

SAI MODEL

H. van Dop

The next model is the SAI model. The core of the model is Eulerian. It contains more than one layer and it has vertical resolution, parameters that are essential for regional oxidant modeling. It is suitable for use on a regional scale, that is, a scale of 1,000 km x 1,000 km. The grid size is limited not only by the intrinsic model properties but also by the availability of data. The meteorological data are in fact critical in this sense. We think that the grid size should be as small as possible. It depends on where the regional application is. For the United States, the size would be of little use if it were less than 50 km, because there would no longer be any resolution of the meteorological data.

The model is to be used in the development of control strategies. Therefore, I think that the model should discriminate between high and low sources. I think the SAI model fulfills this requirement.

As for the meteorology, the meteorology put into the SAI model, as with the other Eulerian models that were presented this morning, is to be prescribed more or less. This is an important point. If it is not prescribed or contained by the model construction itself, the model cannot be used in an application. The performance of a dispersion model can only be judged together with the model input fields. I think it is essential to prescribe how meteorological data are obtained and processed for these models. Without that, the model is incomplete.

Another point is the availability of meteorological data. The availability of meteorological data depends on the area concerned. In the U.S. and Canada, the density of the meteorological network is more or less critical. The data are too sparse in most regions to construct a detailed windfield and turbulence, which are required for this model. In large parts of Europe, the meteorological data are sufficiently dense in time and in space.

As for the chemistry, there was no consensus on the chemical submodel to be used. In fact, we had the same problem with chemistry as we had with meteorology. The modeler does not prescribe a particular system; he/she makes a recommendation. If the user does not like the recommendation, the modeler is willing to replace it. Like the problem of meteorological data, I think this is important. In my opinion, a photochemical oxidant dispersion model should contain both routines for chemistry and preparation of meteorological fields.

A few chemical models were discussed and the consensus was that there is not one good model and one bad model. They all give different results. A study could perhaps provide some answers as to which chemical subsystem should be recommended for use in regional oxidant modeling.

A separate discussion developed on the individual grid scale phenomena mentioned by Robert Lamb. Nobody knows exactly what the effects of subgrid concentration fluctuation will be. It presumably plays an important role, and it should be looked into further.

As for emissions inventories, the mesh size for emissions inventories should not be too small and should be consistent with other mesh sizes used elsewhere, for instance 50 km x 50 km or somewhat less.

I would like to make a separate point for Europe and the U.S. In Europe, a large emissions inventory, the OECD emissions inventory, exists. It is, in our opinion, too coarse for regional oxidant modeling. It consists of grid cells of 127 km x 127 km. This is too coarse for photochemical modeling studies. A large number of emissions inventories exist on smaller scales, on urban scales or on scales far less than 1,000 km x 1,000 km, which could be integrated for regional modeling, but that is not yet the case. In general, these small-scale emissions inventories are of fairly good quality. They contain a lot of components and fine resolution, and they are categorized accordingly. So, the quality is not too bad. The problem is the integration of these inventories.

In the U.S., there is at least one data base on the right scale (1,000 km x 1,000 km). I am not quite sure whether it contains all the necessary components such as HCs.

Further, it was felt that a good emissions inventory should also contain background emissions, that is biogenic and natural emissions from outside the area.

As for the operational properties, the SAI model is operational now. It is "validated," which means the model has been compared with data. Some correlations and statistical comparisons have been made, resulting in a satisfactory correlation coefficient of approximately 0.7.

Our discussion indicated that it might be a good idea, as an extra aid in validating a model, to have a panel of experts judge all the "ins-and-outs" of a particular model. The panel could then come to a consensus such as: "This model meets the present standard of knowledge and does not contain erroneous assumptions. As far as we know, it can be applied for the specified purpose." To have such a panel review a model might be a useful institution in the validation process of modeling.

DISCUSSION

S. Reynolds: I would just like to expand a couple of points that you made. The first concerns meteorological input procedures and the idea that there is a lack of consensus regarding a procedure or specific recommendation. We have had occasion to apply the model in the U.S., and the procedures that we used with the available data base in the U.S. for preparing the complete set of meteorological inputs employed in the model do exist and reside with the model. Any time you consider applying a model in a new situation, you need to look at the available data and the prevailing phenomena in that area and find the procedures that best match the data and the phenomena, the procedures that will give an adequate representation, interpretation, of the data in light of the phenomena that you are trying to represent.

So, on the one hand you have procedures that are with the model. On the other hand, you wish to scrutinize those procedures in terms of any new application to see if they are indeed adequate, to determine whether other procedures might be better in that particular setting.

With regard to chemistry mechanisms, we could make a recommendation on how the chemistry should be treated. There are also other investigators who are dealing with chemistry, who have other opinions on how chemistry might be treated, just as meteorologists have different opinions as to how certain meteorological phenomena should be handled. So, on the one hand we have suggestions as to how the chemistry should be treated, and these are embedded in the model. On the other hand, we recognize that other people have their own opinions as to how these should be handled. Chemistry packages can be interchanged in the models or modified; they are not fixed in time.

J. Killus: To add to the comment concerning chemistry, we have specifically designed the current chemistry module in the SAI model to be both computationally efficient and consistent with the current knowledge of photochemical kinetics. If you wanted to replace that module with some other chemistry, which in fact has been done other for test purposes, emission studies, and other applications, some small additional effort would be required to replace the module. However, in our view such an effort would result in a considerable increase in the computational time required by the model. The specific design criteria of these chemistry data are computationally efficient. So, our recommendation for the chemical kinetics is perhaps considerably stronger than our recommendations for the various meteorological input preparation procedures.

E. Runca: I would like to have some further clarification on the way the model describes the vertical processes in the atmosphere.

S. Reynolds: I am not sure I understand the question.

E. Runca: I would like to know how the model is treating vertical processes in the boundary layer, especially exchanges between the mixing layer and the layer above the mixing layer, and how the model is treating the processes that take place during the night.

S. Reynolds: If you recall, there are 2-1/2 layers treated. Starting from the top, there is an inversion layer; under that, there is a mixed layer and a parameterized surface layer near the ground. We do consider exchange between the layers as accounting for large-scale convergence-divergence effects, entrainment, or detrainment, as layers rise and fall and as transport removal occurs at the surface. Within the mixed layer and the inversion layer, however, the pollutants are assumed to be well mixed.

H. van Dop: That is why I brought the point up here, because the interface of these layers is meteorologically determined. Since you mentioned this relationship, it is essential to know how the layers behave in time and space and how they are modeled.

E. Runca: Is the behavior in time and space of these layers computed in some standard way in the model or is it an input to the model?

S. Reynolds: Well, it is completely determined by data inputs. The model does not assume any particular spatial or temporal behavior. It is coming from the data.

R. Lamb: I understood you to say that the grid size of the model should be smaller than the resolution of the meteorological layers. Is that correct?

H. van Dop: Yes.

R. Lamb: I disagree with that. In this problem and the problem with oxidants, two things are in effect going on, chemistry and transport diffusion. If you have a limited amount of meteorological data, you have to treat the subgrid scale flow variations and a diffusion. The other part of the chemistry, the chemical reaction, the rate at which the reactions are going on, depends on whether the materials have mixed and the amount of segregation of the materials.

H. van Dop: Of course, but the chemistry is a molecular process. To describe the mixing of contaminants down to that scale, you would need a mesh size of a few millimeters, rather than 10 km or 20 km.

R. Lamb: No, I am saying that the resolution of the concentration model should be comparable to the spatial variations in the emission distribution. If the emissions are isolated in various pockets and the pockets are separated by a distance smaller than the resolution of the meteorology, you in effect premix all the emissions from all these sources in the model because you lump them all into one cell. For reactive materials such as O_3, if you alter the NO_x and HC concentrations in as similar fashion, you are talking about large errors in the prediction of O_3.

So, we are right back to the subgrid issue. In my judgment, the necessary resolution of the concentration model should also depend on the spatial variations in the emissions for the region of interest.

H. van Dop: How long would it take before it will be mixed in boxes of 20 km x 20 km, for example?

R. Lamb: Although the chemistry and the mixing are coupled to some extent, imagine a case in which you have two isolated sources within one grid cell. The wind is blowing in a direction that makes the two plumes parallel to each other. They are not mixing at all. However, when you put the emissions in the model, you mix them because you put in all the emissions. You premix them when, in fact, they are not mixed. So, the total O_3 in the two plumes is "x" concentration of O_3 in each of the two parallel plumes. If you mix the emissions, you get an area average concentration that is less than, probably more, than that.

H. van Dop: This discussion is perhaps somewhat technical. However, if you have a windfield with a resolution of that same 20 km and if you have a variation in flow direction with height, it is easy to get a deviation from the mean flow direction of 20° from hour to hour or from level to level. Consequently, the flow field will be such that, due to shear effects, complete mixing will occur within a few hours.

R. Lamb: During convection in the daytime, there is not much shear in the mixed layer. So, plumes tend to be long and slender.

H. van Dop: That is right.

R. Lamb: Some of the data that Henry Cole showed us for the New York area indicate that there are quite narrow plumes, even from those cities. The contours of O_3 are quite high. So, if you lumped all the emissions in that area into one large cell, you would probably not predict O_3 levels comparable to what they saw.

H. van Dop: What would you have tried as grid size?

D. Jost: Just to stay informed of what you are discussing, is there a subgrid mechanism in this model? The subgrid may be for chemistry and for transport, but mainly for chemistry.

R. Lamb: It is mainly for the chemistry. I am saying that the subgrid scale problem increases in magnitude as the grid size increases. There is no good way of parameterizing that in general. The only way to mitigate the problem is to make the grid size small.

S. Reynolds: I agree with Dr. Lamb regarding the resolution of emissions. If we look at other applications for photochemical models on the urban scale, meteorological data aloft are relatively sparse. Yet, we tend to use relatively fine emissions grids. You might push for finer grid resolution, which would enable the discrimination between sources a bit better. So, from intended applications you can perhaps see a little more in detail the characteristics in the concentration field, but one is not looking to go very fine in scale. We are perhaps talking about factors for two choices of grid size.

F. Vukovich: In theory, I think Bob Lamb is right. In practice, however, a large problem occurs when you take meteorological data from stations that are 100 mi apart and try to apply them on a grid spacing of 18 to 20 km. I think Bob would be the first one to agree that you are going to produce all kinds of errors in wind field by some kind of interpolation-extrapolation techniques in developing a wind field that you would use in a transport model. If you look at some of Ed Lorenz' work on predictability, you will find that tremendous errors occur in your prediction field an hour after you have started predicting because you have a misguided wind field. So what you have is a problem of theory versus practice. It also depends on how you want to use your model. If you want to use it as a diagnostic model, in terms of performing control strategies, then a

20-km grid is all right. However, if you are going to make real-time predictions with your model, then you have problems. A 100-mi station variation between wind measurments, bringing it down to 20 km, is going to produce all kinds of errors.

J. Killus: I might amplify Bob's statement by pointing out another phenomenon that occurs during chemical reaction. There is a time constant associated with various chemical processes in O_3 formation, which is fairly long, several hours. So, phenomena that occur on a shorter time scale, less than several hours, are often more or less damped out by the time the O_3 begins to build up through the day. That does not mean that subgrid scale of less than 80 km do not occur. In our simulation with 80-km grid cells, it was quite clear in some of these cases where the station observation diverged from the model predictions. In fact, there were some processes smaller than 80 km operating. Our solution to that is in fact inherent in what we designed the model for, which was to generate a large-scale O_3 flux for, among other things, specification of boundary conditions for much smaller and finer resolved models. In that situation, if you had your two plumes that perhaps would not mix at the smaller resolution, we would suggest that you apply a much more detailed model in the grid. In that case, we have a specific situation that we have again offered in the Philadelphia example, where we used an urban model with a resolution of 5 km, much less than what we are talking about on a regional scale, but whose boundary conditions were prescribed by a model that had a resolution of 100 km. The Philadelphia results indicated that those local emissions generated local concentration patterns. However, the regional transport from, say the New York metropolitan area, perhaps only needed to be resolved at the 100-km level.

HOV MODEL

E. Runca

As for the Hov Lagrangian model, we did not try to explicitly answer all of the questions posed by the workshop objectives. We developed the discussion in such a way to keep these questions under consideration, but we tried mostly to identify the fundamental characteristics of the model, the applicability of the model, and further actions that could be taken in order to make the model operational.

In terms of the characteristics of the model, there are two fundamental assumptions, which derive mostly from the fact that the model is Lagrangian and receptor oriented. The model assumes no lateral diffusion and it is a one-layer model. Therefore, there is no vertical resolution, and the concentration of the advected pollutants is considered uniform in the layer. This assumption makes the model simple on the one hand. On the other hand, it might considerably affect the model's results.

We were not able to quantify this effect. There was a very strong recommendation that the model be extended to include the contribution of the pollutants that diffuse and are advected into the above mixing layer. This implies something that I will leave to the people responsible for the model. I think this problem can be solved in different ways. The question is whether one of these ways will prove to be an effective description of the real processes.

The discussion of the model's characteristics focused on the way the model treats the chemistry. Our conclusion is that the model can be applied with any chemistry. Indeed, the chemistry is an interchangeable component of the model.

We also discussed which chemistry could be recommended. Since the model can treat different chemical schemes, the model could be run with different schemes to see what results are obtained by applying these selected schemes. However, we recommend that a scheme be found that reduces the number of species considered and at the same time provides realistic results. On this point, maybe some further comments could be obtained from Gary Whitten, a panel member who strongly supported this suggestion.

Finally, our discussion focused on the model's applicability. In this context, we directed our attention to the temporal resolution of the model. Not too much discussion occurred on the spatial resolution.

The model, as I said, assumes no lateral diffusion. This assumption requires that the grid size be sufficiently large. The grid size that is now used in the model is 150 km. We were not really able to evaluate the effect of this assumption on the model's results.

The discussion focused on the temporal resolution, and we did not reach a firm conclusion on this point. Some further studies should be made in order to establish whether the model can properly simulate hourly average concentrations.

There was some general consensus that the model may be able to provide better results if the time, the length of the averaging time, is extended. Considering these aspects, it is questionable at this point what conclusions can be reached on the application of the model for the development and evaluation of control strategies. However, some remarks can be made in this regard.

First, the model is a simple model in some way. Therefore, it can be used to analyze different options. At this stage, we do not expect that the model can really provide a detailed diagnosis of the impact of adopting different control strategies. However, it can provide some insight on control strategies. One point that should be made in this regard is that the model is receptor oriented and is a Lagrangian type. Therefore, it is in principle more suitable for analyzing episodes than it is for analyzing the large-scale effects of control strategies.

To evaluate the concentration of oxidants in a given point, we have to evaluate trajectories that have reached that point. Therefore, we can evaluate the effect of strategies on that specific point. If we want to understand the effect of control strategies on a region, we have to run the model for many points in the region. We also have to run the model for different time periods.

At this point, it becomes questionable whether it would not be more appropriate to apply an Eulerian model. The panel tended to agree that, for the analysis of the large-scale impact of control strategies applied on a regional scale, the Eulerian and Lagrangian models might be compared.

The final issue we discussed was what further action should be taken to progress with this model, to make the model operational. The model is now available, at least the core of the model. However, a detailed sensitivity analysis is necessary to evaluate the effect of the parameterization scheme that the model adopts on the model results, that is, the assumptions the model has adopted in relation to the treatment of clouds and other processes relevant to the formation of oxidants.

Some priority should also be given to a validation study. Such a study obviously requires data, and there are not sufficient monitoring data for oxidants all over Europe to enable a model validation on the European scale. However, a validation study can be undertaken by utilizing existing data sets and also by performing field experiments.

For example, as far as the chemistry is concerned, smog chamber data are now available from many different laboratories that could be utilized to get an understanding of the chemistry model that is more suitable for this model.

Also, in order to evaluate some of the assumptions connected with the Lagrangian treatment of the advection of the air parcel, it seems necessary to perform some trajectory, to perform some field experiments to characterize the Lagrangian advection of pollutants in Europe.

As for the next step, the application of the model, suitable emissions data should be prepared in parallel with the sensitivity analysis and validation. Considering the resolution of the model in space, the spatial resolution of the emissions inventory should be approximately 100 km. Since Eastern countries are major contributors to oxidant formation during meteorological situations occurring in Europe, the domain of the emissions inventory should be extended to include these Eastern countries. If that is not possible, the frequency of these conditions and the relevance of the meteorological conditions affected by Eastern emissions should be evaluated before a program of model implementation. Finally, we recommend that natural emissions be considered. In relation to the evaluation of HC emissions, we recommend that some agreement be found between the countries in defining emission factors.

I would like to conclude with a personal comment. It seems to me that a problem of this size requires the coordinated effort of several European institutions, and I would like to recommend that more emphasis be placed on the validation of the model than on the application of the model.

DISCUSSION

R. van Aalst: We pointed out that a detailed chemistry treatment of HCs is probably not quite suited to the detailed HC data in Europe. So, we pointed out the lack of that as well. And about that one of the main reasons for reducing the complexity of HCs.

A. Eliassen: Regarding Dr. Runca's presentation, we pointed out, first, that we have no lateral diffusion, depending on travel time obviously. We have the first, instantaneous diffusion, the grid size, but there are certain problems with Lagrangian models that are more or less unavoidable. One of these problems is that it is very difficult to have lateral diffusion, depending on the travel time and the nonlinear chemistry. If you think about it, you will find out that this is more or less impossible. So, this is a property of the approach that we are taking, this should not be concealed in these discussions.

On the other hand, one might argue that to go to a grid size smaller than 150 km is questionable if you account for two things. One is the availability of the emissions data and the quality they have impressed in the meteorology. The other is the uncertainty of the trajectory calculations. If you follow a trajectory backwards for 4 days, the uncertainty is considerably larger than for 150 km.

The possibility of making the model suitable for calculating hourly concentrations, for example, is rather small, because you need rather sophisticated meteorology to calculate hourly concentrations. One would like to have wind shear together with, for example, the diffusion for such effects. The possibility of doing that with this approach is not good.

If you use a very short time scale, you have to calculate the degree of trajectory that arrives very often at the receptor point. The computer time required then approaches that of an Eulerian model. This also happens if you use very long trajectories, because you do very nearly the same calculations many times following nearby trajectories. Then, the advantages of the Lagrangian approach disappear.

As for the advantages in this calculation, you can have almost any chemistry scheme that you can think of, and it can be run for quite a long time. We can cover a much larger area than we are running at the present.

My last comment is on the need to have an exchange between the boundary layer and, say, the free troposphere. This is also difficult because these have different velocities on the boundary layer, and you get into trouble. It is not impossible to solve, but it is rather cumbersome. It is much easier if you can treat the troposphere as a very large reservoir with constant concentrations. Then you can perhaps do some sensitive things.

E. Runca: I do not find any contradiction with what has been said, unless I misunderstood your comment.

A. Venkatram: I would like to follow up your comment on validation. You said we should do validation. We really do not know how to validate the model, do we? It is very subjective at this stage. Maybe we should have some discussion on the procedure or something of that sort. My personal feeling is that the experts agree that you do not have to rely on statistics to determine whether the model is good. We have been talking about a lot of models, but we have forgotten that a lot of experience with oxidant modeling has been acquired over the past 10 yr. Is there any hard evidence to suggest that these models do not work? If they do not, have we identified the physical mechanisms that give them errors?

E. Runca: The answers to your questions really require a discussion from the audience. However, as my last recommendation, which is a personal recommendation, I really believe that derives from the presentation and the discussion we have had during the previous days. Models appear to be in the research stage rather than in the application stage. To progress further, the coordination of different groups of researchers is required in such a way that a clearer strategy for the next step of model development and application can be identified. This means, in my view, validating which types of modeling approaches, according to various operational needs, should be applied.

G. Whitten: I would like to clarify a couple of points on things that I have recommended in the chemistry. As for the chemical approach, you can take either a broad approach, where you follow many primary compounds, or a more in-depth approach, where you follow fewer compounds but where you follow them further through their oxidation steps. The present chemical mechanism in the Hov model is a broad approach. It follows very many primary species, but the chemical steps are very short and some are left out. There are approaches where you have fewer species but greater depth to them.

In evaluation procedures, a chemical mechanism can be validated out of the atmosphere in a data base that exists in smog chambers, and this is a recommendation that you pointed out. I think that another word in place of "validation" could be used at this point for the atmospheric models. Perhaps you could say that it is time for a "dress rehearsal," to go out and use the models as they now exist with the data base that now exists, to find out what is missing. These models would not be used at the present time where you would expect any regulations or control strategies that were performed to be corrected. However, we need to find out how well the models can perform and, if they are not performing, what is missing.

J. Bottenheim: I strongly agree with what Venkatram says and with what Gary says too, but I suspect that if we start to validate, every modeler will accommodate his own model in his own way. In particular, if a validation shows that the modelers results do not match his validation set, he will come up with fudge factors for which there is more or less a physical reasoning and the modeler will thereby improve the outcome of his model. So, if we get into this validation business, it is very important to have a panel or an independent review committee to assure that the validation is a realistic validation and not a modeler just propping up his own model.

Secondly, I would like to ask Venkatram which models over the last 10 yr worked, because I think that almost all oxidant models are urban models.

A. Venkatram: We, first, haven't decided by what means. My understanding is that we know enough about the chemistry, that there is no substantial difference between the chemical modules. That is one important component of the oxidant model.

The second point is we have been making a lot of points about subgrid-scale chemistry and other phenomena. Is there any evidence to suggest that these will really screw up the results? Hard evidence?

E. Runca: Maybe I should explain myself. When I say that the validation program should be a coordinated effort, I am implying that the validation should not be just an exercise of running the model and comparing the results with some measured data. It is just a problem of research coordinated between different institutes with different expertise to assess which processes are important and what we can say in relation to some of the issues mentioned so far. I really think that this should have a strong priority in Europe before moving to the implementation and application phase.

D. Jost: I too got the impression that it is not as easy as it may be inferred from your comment. Even the different chemistry models are so different that you may come up with very different results with such models. Therefore, we are not at the stage where we can take what is available and rely on it.

G. Whitten: One problem that exists with the chemistry modules right now is that, in many cases, a given data set can be simulated with several different chemistry modules. However, if you then apply control strategies using different chemistry modules, they predict different control strategies.

The research that is now underway is investigating why one chemical mechanism gives different control strategies, and we are at the stage of arguing about parts of the chemical mechanism that lead to different control strategies. So, the fact that you may validate on a given data set is one thing, but when you apply the control strategies that these models will be used for, you produce a new problem.

J. Bottenheim: One case in point is trying to validate a chemical mechanism through smoke chambers. Everybody knows that smoke chambers have problems, and different modelers use different chemical mechanisms, different fudge factors, to take into account what the smoke chamber does to the chemistry. And all the chemical models work pretty well! But, have all the chemical models really been tested from the same smoke chambers with the same fudge factor?

J. Killus: I want to explain what Gary was describing and disagree with Jan Bottonheim somewhat. There are definitely certain chemical mechanisms which, when applied to smog chambers or certain sets of smog chamber data, do not replicate the smog chamber data, no matter what fudge factor you use. We have various examples of that, and that is part of what Gary was saying when we try to locate what specific features of kinetic mechanisms are responsible for these differences in control strategies.

Part of the problem is that kinetic mechanisms have been in a state of intensive development for the past 10 to 15 yr. The lead time necessary for publication of these mechanisms is such that, by the time the mechanism is published, it is more or less obsolete. Mechanisms that are available to people who do not actually design and develop them are usually 4 or 5 yr out of date and may contain certain known errors in fundamental chemistry that have been corrected in more recent mechanisms, errors that might produce some differences in control strategies.

We have found that the differences in control strategies are in fact much less than one would expect from the differences in fundamental chemistry, that is, mechanisms that have even fairly large errors can still often predict similar control strategies within a factor of 20% or 25% of other mechanisms. That 25% difference in HC control can, of course, mean many hundreds of millions of dollars, which is not an irresponsible issue. However, the difference is similar or much smaller to the differences that are going to occur in atmospheric modeling methodology.

In agreement with Venkatram, I would say that most of the mechanisms of recent vintage are reasonable. Properly applied in the atmosphere, they will give more or less proper results. The difficulty is proper application. The various assumptions that one must make in applying these for emission inventories, which may be improperly speciated or may not be speciated at all, will overwhelm the differences in kinetic mechanisms.

My own feeling is that more of a difference exists in kinetic mechanisms in their application state than in the actual fundamental chemistry. Some mechanisms are much easier to use. Some of the parameters associated with these mechanisms are much easier to specify, and that is perhaps what Gary was talking about.

LAMB/NOVAK MODEL

F.B. Smith

Panel III discussed Bob Lamb's and Joan Novak's U.S. EPA model. To summarize the basic nature of the model, it is an Eulerian-type model with a grid length of about 18 km. There might be a good case for reducing this grid length to about 9 km, half of the present size, in order to develop a model that can be used for some regional-scale and also urban-scale modeling.

In the vertical, there are four layers. The model does include a surface layer, in which all of the interesting initial chemistry occurs. Mean vertical motions are incorporated in the model, and these are derived from the horizontal divergences of the windfield. There is also vertical and horizontal diffusion. The horizontal diffusion takes into account the stability and depth of the mixing layer in the usual way.

Advection depends upon the measured winds at the surrounding meteorological stations. As Bob described, this is one area where there is a certain amount of uncertainty, i.e., various trajectories can be derived according to the different ways in which you interpolate between the measured winds and according to the forms of advection.

We might want to discuss afterwards whether you might do better by incorporating some sort of mesoscale meteorological model, into this whole system. Of course, it would enlarge the model considerably; nevertheless, it might give you a better feeling for the advecting winds.

A variable mixing depth is incorporated. Additionally, there is an actual treatment for convective cloud. Cloud plays a very important role. If it is sitting on the top of the mixing layer, various pollutants within the mixing layer can be advected into the cloud, undergo chemistry there, and come out again into the mixing layer. It can thus be a very important part of the whole system.

The model also recognizes the effects of the surface terrain. The terrain has an effect by producing slow vertical velocities through the divergences of the wind field. It also affects the dry deposition of the various substances.

Because the model is directed towards rather extreme situations in which precipitation does not likely occur, there is presently no provision for representing the effects of wet removal by washout and rainout.

As Bob mentioned earlier, there are problems in representing the chemistry going on within the individual plumes on a subgrid scale. Roughly 80% of the computer time is involved in working out the chemical processes, and only 20% is involved in the meteorology, the advection, etc.

As for the background to the model, which we discussed in our panel, the U.S. has decided upon certain air quality standards for oxidants, certain maximum 1-h values that should not be exceeded. They have also developed possible strategies for emission control. The aim is to find a model, such as this one, that can determine by 1987 which of the various control strategies is the optium one to apply. As we have already seen, the model involves the "best available" physics and chemistry as it is presently known. Its modular form is very developed; as our understanding of various processes improves, we can change the various modules.

One very important aspect of the model is that it predicts the concentration fields, recognizing the uncertainties in various processes, the uncertainties in determining wind field, boundary layer height, etc. Because of these uncertainties, a statistical aspect is introduced into the model. Thus, the outcome is not just a single predicted concentration but a range of concentrations with certain probabilities associated with them.

This is a very important, interesting, and novel aspect, which will enable us to look at the model's sensitivity to possible errors. In addition, it will enable us to decide the likely accuracy and certainty with which you can make predictions and the possible certainty with which control strategies can be applied. I think this is a very interesting and useful aspect of the model.

As to the time schedule, the model has been coded and is essentially ready to run. There will be some computer runs this summer. The emissions inventory is being developed under contract and should be available at the end of this year. When that is available, Bob's team can run the model to see how it operates in practice.

Next year, the model will be run under the same conditions as those that occurred during two monthly field experiments in 1979 and 1980, when a great deal of surface data were collected by balloon and by aircraft. The model will be tested for these two monthly periods, and certain developments will obviously result from this test. Having attained the second-generation stage for the model, various proposed strategies will then be investigated in the following 2 yr.

The panel next discussed the appropriateness of the Lamb/Novak model for application to OECD Europe. The first thing that came out of this discussion was that Bob's team really does not have the capacity to revise the model or apply the model to the European scene. To do so, some team in Europe would presumably have to take the model over, refine it or revise it, find its wind fields, etc.

In doing this, the team would be faced with the problem of emissions data. As we have heard from Anton Eliassen and others, the emission situation in Europe is not as good as it is in the United States. which brings us to the problem of boundaries. One aspect of applying the model in the United States is that most of the emissions, certainly as far as the Eastern United States is concerned, are concentrated in the northeastern corridor. As a result, it is possible to choose boundaries, as Bob has done, sufficiently far away from the main emission areas so that, by using the measured surface concentrations in a relatively simple way, you can estimate the influxes across the boundary without incurring any great errors in the whole process. The same is not necessarily true in the

European situation where the industrial belt runs from Central U.K. across Northern France, Belgium, Holland, through the Ruhr, to East Germany, and Poland, going east-west and covering a very large area. Consequently, there are no really convenient boundaries to choose in Europe in contrast to the situation in the U.S.

Also, the model is a very complex model. It is not intended for daily operational use. It is really intended for testing and developing strategies. This brings up several questions. What are we trying to do in Europe? What are our objectives? Are they the same as in the U.S.? Are we going to consider certain strategies for limiting oxidant levels or will there be other objectives? If there are other objectives, this will affect the sort of model that we will need to apply. If we are going to use the model in Europe, we will really need an institute in Europe, either an existing institute or a newly created one where a lot of effort could be devoted to a model of this kind, not only to the model itself but also to the whole backup behind the model--what are the objectives, how do you define them, what are the objectives based on, and so on. So, I recommend that OECD consider setting up an institute for this sort of work.

As an alternative, if we do not want to use a model of this kind and of this complexity in Europe--if we want to develop our own simpler model--a model of this kind could at least be used as a standard by which to test a simpler model. Maybe we could take up the offer Basil made at the beginning of this meeting. Maybe somebody from this European institute could come along and test the model side-by-side with the selected U.S. complex model.

Finally, coming back to the point of what our aims in Europe, we have to identify the actual hazards that we are particularly concerned with in Europe. Are they hazards to health, to vegetation? What are the time scales involved? Is it a question of an hourly oxidant level, a daily oxidant level, or an annual level? Having decided upon the levels and time scales, which model is the most appropriate for this sort of investigation?

DISCUSSION

S. Reynolds: When you state that the model provides an estimate of the uncertainty, is that a direct calculation or the result of performing many simulations in a sensitivity mode?

F. Smith: As I understand, it involves repeated running of the model using different assumptions for some of the factors in the model. Is that right, Bob?

R. Lamb: It is primarily associated with the specification of flow fields, which gets back to our earlier discussion about the resolution of meteorological data. It is a rather complicated thing to describe in a few words. Essentially, the flow fields cannot be described uniquely. There is a set of possible flows, that is, a set of flows that are possible within the constraints of all the observations that we have in space and time and for physical transport. Within all of these constraints, there are many possible flows.

We can assign probability for each of these flows on the basis of empirical spectral data that have been seen for the flow field. In essence, the uncertainty exists mainly in looking at the nonuniqueness of the flow field. The uncertainty is associated with that and not so much the uncertainty due to errors in emissions or errors in known factors. There are separate problems.

A. Christie: How much confidence do you have in the kind of data used to indicate the degree of uncertainty, the probability of having any particular flow distribution field? You have a spectrum of possibilities.

R. Lamb: Right.

A. Christie: Now, you say that you are going to do this from an analysis of information on the flow fields. Do you feel that there is enough information at the present time to carry out this analysis and that you would have the same confidence in the flow analysis across North America as a whole, or would these flow patterns incorporate mesoscale phenomena and terrain effects? Do the data exist over the model domain to adequately define the probability of the flows at all locations?

R. Lamb: With any given, the uncertainties in the flow would be very large, even if there is only one source of meterological data, only one station. Theoretically, if you are given a set of observations in space and time and if you are given the physical principles that are used in the mesoscale flow model--momentum, energy, mass conservations--you can delineate a set of flow fields to whatever resolution you wish. There is a set of flow fields, and each field is consistent with that information. Each of them obeys continuity, momentum conservation, all the observations you have. Within that set of possible flows, you can then take information such as the climatological data that are used to get optimal interpolation formulas where you find weighting functions that minimize in some climatological sense mean square error. We can use climatological data or historical meteorological data as the empirical basis on which to assign probability to the flows in this set. Some of these flows may have no probability or vanishingly small probabilities on the basis of what we have observed from the atmosphere in each individual region over long periods of time. In our case, the empirical data base would then be used over about a 2-day period, where we have observations and space and time. We review these empirical data to assign probabilities to all the flows that are possible during that period, given the physical constraints.

In theory, we have worked this out and we are trying to implement it. We have made some progress, but we have quite a way to go before we can look at just how large the uncertainties are.

G. Whitten: It is not clear to me why some of the uncertainties that need to be investigated, that are being built into this model, cannot be investigated with existing models right now. For instance, uncertainties in the flow field could be, and their effect could be tested now with other models to see how important that would be when the Lamb model is ready.

Another thing is the microscale mixing effects of parallel plumes. If you use an existing model with a very fine mesh size in an area, you could make hypothetical parallel fields and investigate the effects with existing models. If they are found to be extemely important, they would have to be built into the model when it is ready.

I do not see the necessity of waiting for this model to be ready and then tested when a lot of these tests could be done at the present time.

D. Jost: As I understand it, there are some available models that are in some respects very detailed. One would be able to check the sensitivity relative to simulations with those models.

R. Lamb: It is not quite as simple as that. In testing the uncertainty due to atmospheric flow, you can certainly take any model, drive it with different flow fields, and look at the differences you get in the prediction. The problem is defining a set of flow fields that is consistent with all you know. Unless you are testing with the proper set of flow fields, the results you are getting will not be applicable.

As to the effects of the grid resolution, the subgrid, could be tested to some extent. However, tests for flow field uncertainty, which is in our case a major consideration, will have to wait until we have developed a way of defining flow fields and assigning them probabilities.

G. Whitten: The sensitivity to the flow fields could be tested. They might be incorrect flow fields, but you could determine the sensitivity to them.

R. Lamb: Yes, but you may be using such an extravagant range of flow fields that your results are very pessimistic, whereas in reality the situation is not that bad. Or, you can take a flow field and not disturb them. There is an entire range of variations of flow fields that you can put in and get a very wide range of answers. If you can define that set of flow fields in advance, then, yes, you can do that.

E. Runca: I am not questioning the reality; I am only commenting. I think that this group made an important point, the need for a European institute for a study of this type. I support that idea. We now recognize that many environmental problems in Europe are not local, that they are problems for the whole of Europe. It might be worthwhile to devote more attention to the possibility of creating an institute to deal with these problems.

A. Venkatram: I would like to follow up on Bob's remarks about specifying the velocity for looking at the sensitivity to concentration predictions to the unresolved components of velocity. We are doing some preliminary studies on that.

Instead of spending too much time on that, I would like to point out that if you want to calculate concentration variance, you have to worry about not only this so-called inherent variance, but also the variance associated with the model inputs. You cannot predict what the meteorology will be next year; therefore, your control strategy must account for that variance, which of course means that the total variance is going to overwhelm things. That is going to pale into insignificance. Regulators have to start thinking in terms of probabilities; in fact, it is an educational process.

S. Reynolds: Dr. Lamb, what is your schedule for doing the flow field uncertainty work?

R. Lamb: The project is running somewhat parallel to the model development, and we are hoping to do part of it within the next year. The main problem is this: One often talks about using a mesoscale flow model to drive the concentration models. In such cases, you incur a great amount of error as you initialize the flow field model with the meteorology at some moment and as you use the prediction of that model to go into the future, because the predictions of a mesoscale model do not arbitrarily remain acceptable far into the future. In our case, we are doing a simulation of a historical event. It is a so-called "worse-case meteorology" in the past in which you have observations of what happened for a time interval of 2 days.

In our approach, we have taken the equations and in effect put in all the observations of time and space. We are now defining a set of functions that satisfies all of those observations and all of those equations. Within that set, we can then assign probabilities. This is a very complicated problem. It requires working in high dimensional spaces, and it is a very time-consuming problem for the computer. However, we are proceeding with it because we think it is worth exploring. If it turns out to be impractical, we will just have to face that.

A. Eliassen: It seems to me that you do not really know what sort of probabilities of the front flow fields you will have in the future, say, if you are going to construct control strategies. You will get certain situations and you will assume certain probabilities in those situations according to the flow field. What is going to happen next year and what sort of probabilities will exist at that point is very difficult to say. The degree of the control strategy will greatly depend on the flow functions. Then, I would be very surprised if this is at all really necessary to consider.

R. Lamb: There is some misunderstanding about what I mean by probability. We can discuss it later.

H. van Dop: I would like to remark on what Runca said a few minutes ago. We are talking about regional oxidant modeling in the United States and in Europe. In Europe, no place exists to carry out such studies, and I would like to hear the participants' comments on that topic, because it is now clear to me that only in the United States is there a platform for such studies. That is an important conclusion if it is true.

D. Jost: I think it was presented here in a different way. If we want to apply this model in Europe with all the SAT submodels, it needs a home in Europe that is not yet available.

E. Runca: The question is a little bit more general than the way you are now discussing it. I think we have reached a point where we recognize that in

Europe there are many problems--not only this oxidant problem--which are of interest to Europe and which in many aspects concern the European environment, problems that are essentially multidisciplinary and require the coordination of different groups of experts for their solution.

If we consider, for example, the long-range transport of sulfur, we realize that the problem is extremely complex, because the development of a model that describes this problem has to take into account so many factors, which really requires the contribution of many groups of experts. It requires the synthesis of the findings of these groups. So, there is really a need for a platform in this sense, not only in coordinating, but also in taking these results and doing additional research on these results in order to mesh them and create the possibility for the analysis of results and their application to the evaluation and development of control strategies in Europe.

My statement may be naive in terms of a model for consideration, but it seems that a practical problem to be solved, in order to progress in Europe in dealing with these environmental problems, is to find a home for these types of studies. Sooner or later, we will have to consider the creation of an environmental institute in Europe.

D. Jost: I did not want to get into this; I only wanted to explain what the group meant. We could not propose an European environmental institute. The group's proposal was directed toward the handling of these very models.

A. Galli: Another thing is perhaps not just limited to Bob's model, the Lamb model. I am not sure that it does not exist if they choose to use the SAI model or the Carmichael model.

All of these American models really do not have a home in Europe. Bob's simply came to the front for use over there, but none of them really does.

S. Zwerver: I do not think this is the place to discuss it. We are discussing technical questions of models. This is a problem for European countries. We discuss with each other in international grous, like OECD or. I think that's more of a platform to be decided by an international cooperation, the Europe International Institute, as this here. I think it is good to say that in Europe there is something missing in the technical coordination.

D. Jost: I wasn't going to decide here on the kind of cooperation in Europe, but there is the recommendation for further coordination.

P. Lieben: Just a very short comment here. OECD is quite ready to assist in developments and recommendations on which strategies should be adopted and so on, but in the European context a lot of emissions are impacting the situation in Eastern European countries. We have our normal boundaries in OECD. It is a question of coordination not only among European countries, but also among different international platforms that exist on the European scene.

D. Jost: That is right. One more question, and we will come to the next presentation.

Unidentified Speaker: Did your group discuss computer resource requirements for a typical scenario, a control strategy scenario, using the Lamb model?

F. Smith: We did not actually discuss this in detail. Bob, would you like to comment on that? You actually know.

R. Lamb: We have done some 24-h simulations with the NEDS data, the old emissions data. For one 24-h simulation, the requirement was about 10 h of UNIVAC time. As to how that compares with some other system, perhaps Joan knows.

Unidentified Speaker: I am concerned about the probabilistic aspects of the wind field model. That multiples it?

R. Lamb: Exactly, it is multiplied by a factor of 10.

J. Novak: In terms of the application of the flow fields, we intend to use another computer, which we already have. With that computer, there is a difference of about a factor of 10 in terms of performance, which puts the CPU time for a typical 24-h simulation to around 1 h. This at least becomes affordable in terms of multiple runs, say, the 50 to 100 runs that may be required for a scenario. It is expensive, but you have to decide whether the results you are getting are worth the price.

Unidentified Speaker: Do these model runs involve the preprocessing of the data, the meteorological data base, or are they based on the assumption that preprocessing has already been done?

J. Novak: These figures involve only the model time, not preprocessing time. They are not comparable in terms of preprocessing. Possibly, the flow field generation would be the most time consuming. The others could be several orders of magnitude lower.

R. Lamb: If you consider the regional problem in all its complexity, this problem is inherently one that requires a large amount of computer resources. I mean, it is in the level of the climate modeling work, the large-scale global simulations. We are out of the realm of the old plume model calculation where we speak about seconds of CPU time. This problem has moved up many orders of magnitude into a whole new range of problems if you treat it in this complexity. If you decide that you do not want to pay the price of that and want to do something simpler, then you can do that. The question then becomes: Are the numbers you are getting really reliable enough to base strategy decisions on?

U.K./ADOM MODEL

A. Eliassen

Panel IV discussed two models, the U.K. model, which was developed at Harwell, and the Acid Deposition and Oxidant model (ADOM), which was presented by P.K. Misra. The U.K. model is very complex as far as the chemistry is concerned--40 emitted species and 300 reactions approximately. The meteorology is considerably simpler. It involves instantaneous mixing into a box with the dimensions of 450 km x 360 x 1,300 m in the vertical. It is operational, and it has been used to compare predictions with measurements.

The ADOM model is a multilevel Eulerian model; the North American version has a 127-km horizontal resolution. It has dry and wet chemistry and deposition and nine classes of HCs. It is modular in structure and it is comparable to the Carmichael model. The ADOM model should be operational in 1986, when it will be tested against historical data.

Our discussions concerned the uses, possible uses, and limitations of these two models.

We will present the U.K. model first. In its present version, it does not handle transport at all. Therefore, it cannot be used to develop oxidant control strategies for OECD Europe.

Its complex chemistry might be used as a reference for less sophisticated schemes. However, we felt that it would be necessary before that to test the model against smog chamber data.

In addition, there are plans to further develop the model into a moving cell model. That is a natural extension of this modeling approach, and it would make the model more or less similar to the Norwegian model presented. If this development is done, its complex chemistry would then require an emissions inventory of the 40 emitted species over Europe, and we are far from actually having those data. Also, the work involved in developing the moving cell model would require approximately one person-year.

As to the ADOM model, it is being constructed specifically as a tool for developing control strategies. The model is perhaps tilted slightly more towards acid rain than oxidants, but that may not represent a great problem.

The meteorological input data for this Eulerian model are available in principle for both North America and Europe. The data are more or less sufficient, especially if we cooperate with the National Weather Service and related institutions.

The emissions data are better for North America. In Europe, the situation exists that I mentioned earlier. The quality of the available data is very variable from country to country.

The ADOM model is planned for use in both regions, so it may be of interest to know that, regardless of what OECD decides, the model will more or less be used in both regions anyway. So we could compare, of course, if OECD is choosing another model.

Since the model is still under development, we did not really have a detailed discussion on it. Instead, the group discussed so-called complex models.

To summarize, the simple models are useful for long-term averages, and they often do quite well on those things, for example annual averages. The problem arises when you want to use them for control strategies. Some of their parameterizations are such that it is doubtful whether they respond correctly to assumed emission variations. Thus, even if they calculate averages very well, they are sometimes suspect for use in control strategies.

The complex models are more or less unavoidable. If you are interested in short-term concentrations such as hourly concentrations, the shorter the averaging time, the more complex the model required. Hopefully, these complex models have more correct responses to assumed emission variations. Not being a chemist, I wonder if the chemists' overview of their complex schemes is really correct.

DISCUSSION

S. Reynolds: Could you clarify what you had in mind as a simple model versus a complex model?

A. Eliassen: We more or less regarded the ADOM model as a complex model. The U.K. model is a complex model as far as the chemistry is concerned and a simple model as far as the meteorology is concerned. The same characterization applies perhaps to the Norwegian model.

G. Whitten: I would like to reiterate my comment on the chemistry of the U.K. model, since it is very similar to the Hov model. Namely, it appears to be very complex to a nonchemist. As a chemist, it is far too simplistic to me in that it treats many compounds as primary compounds. The chemistry on each compound is treated rather simplistically, so it lacks in-depth treatment. I would be skeptical of its ability to handle long-term effects, because the chemistry of each individual compound is not treated in much depth.

A. Eliassen: Perhaps another chemist could respond to that.

O. Hov: I am not a chemist, but I think that your comment applies more to our Norwegian model than to the U.K. model, since the chemistry in our model as a transport model is much more simplified than that in the U.K. model. I think that the U.K. model's chemistry was up to date with the current literature at the time of its construction, which was in 1978.

J. Novak: I want to make a general observation. Several of the presentations on the first day made reference to an interest in acid deposition modeling. It might be interesting to do in terms of the ADOM model's ability to accomplish both the acid deposition problem and the oxidant problem.

In terms of the success in this OECD effort, each representative has to carry back to his/her country some kind of argument for funding this effort, some argument that might give extra weight to obtaining support for funding and commitment, because there is an attempt to solve two problems, especially the acidification problem, which seems to be of prime concern in many countries.

A. Eliassen: It would surprise me if OECD once again initiated the program on acid rain.

D. Jost: Perhaps not OECD, but the countries where the problem exists.

N. Laulainen: I'm Nels Laulainen from EPA, the Pacific Northwest Lab. I think OECD is aware of this. There has been a proposal to compare acid rain models--long-range transport models that deal with acid rain--in the future. The U.S. position is that they would like to do this; however, the U.S. would like to have such an effort wait until the completion of this oxidant comparison. For one thing, we can learn something about it. For another, rather extensive model development is occurring in the U.S. now, namely, the development of a new Eulerian acid deposition model. We would like to have this included in such a comparison, and it is not ready at this time.

In summary, we are interested in a comparison with acid rain models, but the comparison would best wait until this is completed and until current development efforts are completed.

G. Whitten: I support Ms. Novak's ADOM approach, combining the acid deposition and oxidant. As a chemist, my understanding is that the acid deposition problem depends very strongly on oxidant chemistry, and there have been a lot of approaches to the acid deposition problem independent of the oxidant problem. The two are much more closely related than approaches have been in the past. I wanted to add my support to that.

N. Laulainen: The work that is being done in developing acid deposition models does not include the type of models that you may be thinking of, the type that tries to just treat sulfur chemistry by itself. The model that is being developed in the acid rain program is quite a comprehensive model that includes oxidant chemistry as well.

G. Whitten: Then that makes it an oxidant model as well.

N. Laulainen: It is, but the model is not ready at this time for comparison, and there are some other models that are being developed that are not ready. We would like to delay until they are ready for comparison.

G. Carmichael: I would just like to point out that this is the approach that we have taken in the development of our model. We are looking at both the oxidants as a part of the regional-scale cycle of trace gases in the atmosphere.

A. Galli: As Dr. Eliassen indicated, the Misra model is going to proceed with or without the support of OECD or the Control Strategies Program, and it is going to be available for comparison. I do not see getting the acid rain situation directly tied in with the control strategies project at the present time, since the acid rain project as a whole is getting tied up in a number of international organizations and is getting coverage there--even though, as far as acid rain is concerned, OECD has perhaps been tied into an evaluation of some of the models.

If we stick to the models before us and the one that is going to proceed with or without our blessings, we need to look at those besides the Misra model that may or may not proceed or receive support. We had a considerable discussion about the acid rain situation in our last AMPG meeting and decided to defer a lot of this, even though oxidant work and acid rain work are very similar in modeling, chemistry, and in a number of other areas such as emission inventories. We may be getting far afield. I can appreciate people wanting to get heavily involved in acid rain, but there are too many people involved in it right now. It is a buzz word now rather than a rationality.

J. Killus: I would like to offer this observation. It is a fairly natural and easy thing at a meeting like this--where you have many technical people approaching a problem, where all of us have an opinion about the various approaches and most of us have fairly strong personalities and strong opinions--to slip into a competitive stance. You know, this model is better than that or let's use this approach rather than that approach, but as this conference has proceeded, most of that competitiveness is, at least in my mind, slipping away. I find nothing exclusionary in these various models or modeling approaches.

There is nothing mutually exclusive about, say the Lamb model, the SAI model, the Hov model, or being able to apply it in Europe. We will use whatever emissions we can get. We will use whatever meteorological characterization we can get, and that will be reflected in the model results.

There is nothing that says that the emissions grid used for the SAI model cannot be used with the Lamb model. We will all be certainly requesting a 20-km emissions grid if you can supply it to us. So, I would just say that I see no reason why we cannot proceed now, and I think that that would probably be a good idea.

R. van Aalst: I would like to make a very trivial comment. I liked a model with simplistic meteorology and simplistic chemistry. If you are going to model the long-range transport of oxidants in Europe and if you start out in a diagnostic way, which would very much be the way to start in Europe, I would advise the use of extremely simple chemistry in terms of modeling oxidant production and oxidant destruction, and just give it a diurnal cycle or something like that and try to relate that to the amount of emissions in a grid. It is just as simple as that to see what is going on and to analyze the results in terms of such simple parameters. Probably nobody proposes such an approach because everybody can do it. I would like to urge that such an approach be taken.

SESSION VI

CONCLUSIONS AND RECOMMENDATIONS

April 14, 1983

CONCLUSIONS AND RECOMMENDATIONS

P. Lieben, Chairman

We are now arriving at the end of the workshop and trying to put everything together. During the next 2-1/2 h, I would like for us to consider what OECD should do after we have reviewed the situation. I would like for us to consider the objective to be achieved, the subjects to be commented on, and the facts that are not known.

The objective is for OECD to recommend to member governments within the near future--not 10 yr from now, but within 2 or 3 yr--regional model(s) that they can use to develop and implement control strategies for chemical oxidants.

I have stated this in very simple terms. I am not an expert, and I am not familiar with all the details that are involved in the chemistry or the meteorology. The question is: What should be done to meet this objective?

First, we will certainly have to assess the effects of oxidants. Do they affect human beings, crops, vegetation, or materials? This question certainly has a bearing on the control strategies that will be chosen by member governments. In Europe, most member governments have not yet decided which control strategies to adopt. They may have some idea, but it is not fixed.

Second, we will have to look at the models and make some kind of selection. This does not imply that some models are good and others are not. We will have to select one, two, or three models that appear to be the best candidates for meeting the objective of developing control strategies. We will have to concentrate on these models, and this will be part of the work plan that we will try to develop. We will certainly have to look at the capabilities of the models, the time frame for availability, the necessary input data, the costs, and possibly other factors. These factors are not meant to be restrictive; they are simply pointers to direct the discussion.

The third item is the development of emissions inventories, especially for Europe where they are largely missing. Emissions inventories will certainly have to include NO_x; they will also have to include HCs, and we will have to get some ideas about the species to track.

One very important point is the grid resolution for such inventories. It will have to fit with the model(s) that are likely to be selected for control strategy purposes. We have heard that getting such inventories is a major effort. I would not like, after 2 yr of effort, to finish with an emissions inventory that is done in such a grid resolution that it cannot be used with the model we finally select.

I think the domain covered by the models that are likely to be selected is also important and will certainly require O_3 and meteorological data. It is probably less important than the emissions inventories.

So, this is a very rough, tentative agenda for developing a work plan. I see these three activities going on in parallel, but converging at some point. In other words, there will be some point where we are ready with the three different elements, where we can put everything together and make a recommendation that constitutes our objective.

Do you agree with structuring the discussion in this fashion this afternoon? I do not think that we will achieve a definite answer on everything now, but we can perhaps get from the meeting as clear an indication as possible of the major elements that have to go into such a work plan.

I should also give you some idea of what is coming in the future. We have a commitment to prepare a work plan for the special session of the OECD Air Management Policy Group in mid-June. We have about half a month to write the plan and to present it at the meeting for discussion with those countries that are willing to go ahead with it, countries that are willing to put forth the necessary effort.

This will be done in mid-June; then we will have the regular meeting. We will have to review the plan in terms of the willingness of member countries to go ahead with it, according to the possibilities they have. In September, we will have the regular meeting of the Air Management Policy Group to finally adopt a plan for implementation.

Are there any questions about the proposed structure for the discussion and the outline for a work plan?

G. Whitten: To add a slightly different complexion to the discussion, we can also consider as part of the plan whether you want to concentrate on a single model, a single type of model, or a battery of models. Our recent experience is that, if you put together an inventory on a grid scale, you can use a grid model, a trajectory model, and a box model from that inventory. Using all three models at once is cost-effective and time-effective in that you learn much about the chemistries, about the adequacy of the speciation and the amounts there, just by running a simple box model. It tells you things very rapidly. So, I am suggesting that a plan of the types of models to use all at once be part of the plan.

P. Lieben: That is a very relevant point. I was expecting some discussion on my point that the selection of models for further work within OECD is understood, that models will progress and that people will go ahead with developing models, etc., but that we have to select for OECD purposes what will suit best. That is certainly something to look at.

Unidentified Speaker: If we are going to get into assessing the various models that have been presented, it may be good to reach an agreement here as to what such a model needs to have inside of it, what it needs to predict, and what time scale it needs in order to qualify as a strategy-development oxidant model. It may be good to determine by what standard a particular model does or does not qualify before you even get into the various models. It seems to me that the various routes may have different criteria within their discussion as to what is an oxidant model for strategy development.

P. Lieben: Yes, I think this will be part of the discussion covered under the second item. What I am trying to do now is establish these three points as areas for discussion and take them one after the other. When we come to the discussion about models, your comment is certainly valid.

E. Runca: Can you specify better "assessment effects of oxidants"? What will this imply? Is it just a review of what has been done so far?

P. Lieben: This leads us to the first point, which I would like Lars Lindau to introduce.

L. Lindau: As has been said many times during this workshop, the time averages are very important. We ask that for the models and also for the discussion of strategies. In talking about health effects, we have 1-h averages. We have both a recommendation from WHO and we have the U.S. EPA standards. Thus, I do not think there is much need for a discussion of the normal time averages when we are talking about health effects.

As for the effects on crops, vegetation, forests, etc., we have short-time averages. To my knowledge, there is also information about the effects over longer time periods for both crops and forests. I have divided these up. Of course, it could be different when you are talking about vegetation. Whereas, when you are talking about forests, you have to talk about the needle life, and that means not just the (summer, half-year); it could mean a couple of years. Needle life is 4 yr.

In a draft proposal made by Swedish scientists about a half a year ago on criteria for determining the effects of chemical oxidants on vegetation, a 1-h average, an 8-h average, a 1-mo average, and a 1/2-yr average were used.

Galli proposed and I am also proposing that we have to assess the effects at the beginning of this project for OECD. OECD and the OECD countries must have a base of knowledge, so they will know which effects they are looking for. This base could be developed in several different ways. Experts could get together to assess the existing literature, or it could be done on a bigger scale. I have not thought the issue over very thoroughly, but there is a lot of information on the effects of chemicals in the literature. If you use scientists from a couple of countries, they could come up with a conclusion, and we could get some information that would be very useful for future work.

P. Lieben: Are there any comments on the issue of effect? This is an important point in establishing priorities and developing control strategies. You have to know what you are supposed to protect and how. Are there any comments on that particular point?

E. Runca: Is there any plan to undertake some projects in Europe to quantify in some way the effects of oxidants on crops, forests, etc.?

P. Lieben: I am not aware of any plan like that. The issue before us now, an assessment of effects, will certainly involve looking at what has been published, what has been put together, and the studies that will have to be made in time to extract a sense of the rest.

In addition to that, will it be necessary to conduct some special studies? I do not know. We do not have any plan to do so in OECD. We are just looking at which plan we should have.

E. Runca: Should we try to make some proposals now?

P. Lieben: I would like to repeat what I said at the beginning. I am not trying to fix anything, but to get some ideas from the meeting of the elements that we can take into account when developing this work plan.

J. Novak: One thing that would need to be determined is how you are going to evaluate the control strategies, and this has something to do with the effects of O_3. If you run these models with different emission scenarios and different control strategies, then a choice has to be made as to which is the best and the most cost-effective model. In order to make that decision, you have to weigh the levels of effectiveness you have with the different control strategies against the cost of implementing them and the costs to humans, vegetation, crops, whatever.

So, it seems that some type of economic analysis would be worthwhile in which a functional relationship is developed between O_3 levels and crop damage or economic loss--however you wanted to define it in terms of the effects of O_3. Thus, given a function of economic loss due to O_3 concentration and the cost of controlling that O_3, you can determine, at least on a more scientific basic, what the optimum control strategies might be in terms of cost and effect.

P. Leiben: Thank you. Are there comments on that point?

B. Thompson: A number of studies were done in the United States on the economic effects of oxidants. Boyce Thompson Institute did several of these studies. They found a strong synergism with other pollutants. So, it is not just oxidants at work; it is oxidants plus the O_2. If you look further, it gets even more complicated than that.

Beyond the first order of effects, it could therefore be a question of ever increasing complexity, and it could end up in a situation in which the acid rain problems are being talked about now. It might be because some pollutant causes some metal ion to mobilize and whatnot. It could turn out to be a Pandora's box.

P. Lieben: It may be.

D. Balsillie: There is a network across the United States called the National Crop Loss Assessment Network, which is systematically trying to find out what the economic loss is on crops across the U.S. It turns out that the crop loss is very substantial. As Joan Novak pointed out, there is nowhere near the amount of crop loss in terms of costs, is nowhere near the billions of dollars it would cost to abate the problem. In place of this, the U.S. raised its standard from 80 ppb to a 120 ppb, knowing that they would have to accept that level of damage.

In Ontario, we are losing about 15 to 20 million dollars worth of crops a year, based on the 1980 averages. However, 15 to 20 million dollars does not come close to the amount of money we would have to spend to reduce the VOC emissions from our two major areas, the Windsor corridor and the Golden Horseshoe around Toronto, Ontario. So, there is going to have to be something more than just the vegetation losses put together. In other words, I think you are going to have to come up with a health effect as well as a vegetation effect, because I do not think that we are going to be able to justify the cost of abatement based on vegetation damage.

A number of people in Europe are working on the effect of O_3 on plants, and they are quite knowledgeable in this area. They have been in the United States on several occasions to take part in meetings such as this, so they are aware of what is happening on both sides of the Atlantic. If you will look into it, you will find that such information is available for Europe. Whether it is actually documented in dollars and cents or some other currency, I do not know.

P. Lieben: As to the assessment of effects on crops, that was never the intention. I think the assessment covers a number of effects in order to judge which one(s) we will try to reduce, and this will determine which control strategies are adopted.

J. Schjoldager: I think the question of plant damage is important. Along with that and as early as possible, we should include in any plan a strong recommendation that countries carry out measurement programs to find out what the concentration levels of oxidants actually are.

Knowing what a slow process it really is from your plan to carry out measurements until you have a program and a good program operational. It is important to urge both countries and research institutions to put this into operation as early as possible.

P. Lieben: Thank you. Are there any more comments on this issue? I know that is not the point that we intended to discuss. I do not think we have to prolong it.

P. Grennfelt: Before we proceed with our discussion of the models, I want to point out that it is essential that the models will present (sounds) that fit into the work to estimate damage.

P. Lieben: I am not sure I understand.

P. Grennfelt: There are many different ways in which biologists use data to estimate effects. We do not know exactly what type of data they really want to get in estimating the effects. I think it is necessary to really get the models that the biologists define what type of data they need so they can put together what would allow this analysis factor to reach abatement. The models discussed until now have very much focused on control strategies without considering the issue of effect.

P. Lieben: Okay, you think at some stage biologists should be brought together with analysts and control people to have them discuss and agree on a way to provide it.

E. Runca: I would like to emphasize this point. These three items--the effects, the model, and the emissions inventory--and the measurement of other data should be the components of a problem. However, these three components should not proceed independently. There should be an overall framework, which is continuously verified. Information from these three components could then be used for the evaluation of control strategies.

S. Zwerver: I get the feeling we are moving to conclusions concerning other fields and fields that we have not discussed in the last two days. We were discussing models.

We have received a suggestion about cost-benefit analysis and effects, and we have just discussed effects as an influence on the models to be selected. So, are we going to select a model for a long average, or are we going to select a model to (produce values)?

I should warn for all these other discussion because they are very complex, and I don't think we have the expertise there to--

P. Lieben: I was going to move on to the second point. If you remember, it concerns the preselection of models or advice about which models are the best candidates to focus on for the next 2 to 3 yr in the OECD program, in view of our objective. I will ask Dieter Jost to introduce that subject, and we will have comments afterwards.

D. Jost: After the discussion this morning, perhaps more aspects more technical and scientific than the models that have been presented here. I think now I (prescribe to some of our discussion panel) the (apparent) recommendation based on this technical and scientific discussion as a result of these discussions and the report of the chairman of this panel.

I would like to describe some of the more administrative aspects of the discussion, which could be a basis for further discussion within OECD. I have taken the models that were discussed and considered some questions on this. I have already tried to answer part of these and I would like to ask you to correct this. You may have additional items that could be taken as the basis for later decisions.

This should include the Hov model, Lamb model, Misra model, SAI model, and Carmichael model. These models are operational to some time, which was mentioned this morning. I did not find out when the Hov model will be operational?

O. Hov: The model has been applied, as published in the Journal of Applied Meteorology, for an episode at the Norwegian southeast coast, and a number of sensitivity studies have been carried out. So, it is operational.

D. Jost: I think this is true. It may have been 1982 or 1983, but it does not matter.

Unidentified Speaker: Is the Carmichael model pretty much running right now?

G. Carmichael: As pointed out, when we discussed the term "operational," we were talking about operational as a verified model to field study. We can compute numbers at this point in time, and it has all the pieces. We plan to apply it to some field study data next year. So, it depends on how you look at the term "operational."

B. Dimitriades: Perhaps we should be asking in what year the model will be turned over to OECD.

D. Jost: By operational, I meant available for people who are not the authors of the models. Should the date be moved to 1986?

G. Carmichael: Yes, 1985-1986. We plan for a users' manual to be available then.

R. van Aalst: I strongly recommend that the Hov model be viewed as a two-layer structure. I do not know whether this should be included in the definition of the operation or condition needed for OECD. Could someone comment on that?

D. Jost: Perhaps I could answer. This takes us to the last point, what we can do with these models. As it now is available, the model is a one-layer model. It could, nevertheless, be used for some tests. This would be the model as it was available already in 1982, without the recommended changes.

Perhaps we could come back to this third point. You can use it for strategies, which is the purpose of this project.

This morning I got the impression that the model would be used for studies in which you want to analyze air quality situations or special effects on oxidant formation. Perhaps this may be changed due to the input data needed for the model. Let's come back to this line a little later.

There have been some estimates on the effort needed for running the models. This was estimated for the Lamb model, but I do not remember the numbers. As I am not a computer specialist, I would like to ask which unit should be taken.

H. van Dop: Make three classes: low, medium, and high.

D. Jost: If I remember correctly, the effort is high for the Lamb model. What is it with respect to the Hov model?

O. Hov: That depends on how many changes are required.

D. Jost: Does that mean high, as it is now?

O. Hov: As it is now, it is low.

D. Jost: Is it medium or high for the Misra model?

P. Misra: High.

D. Jost: The modeler will not be high, but the input. I do not know whether "high" has a different inference in your language. The SAI model?

G. Whitten: Medium.

D. Jost: And the Carmichael model, high. As to evaluation, this is no longer planned since an evaluation has already been done.

A. Christie: However, it has not been evaluated in terms of model resolution, a criticism made this morning.

D. Jost: The model has been evaluated as far as it is available, so the evaluations that you are mentioning are not possible.

P. Lieben: Maybe Mr. Hov has a comment on that.

O. Hov: If the model is to be evaluated according to a set of recommendations made at this meeting, that has not been done of course. The only model evaluation has been a comparison with a measured period of time at one site in southern Norway.

E. Runca: In the final discussion, there was some agreement that studies to evaluate the model are needed. So far, it has been mostly a qualitative analysis comparing with not only one single station. (In terms of describing) the trends of the process, I don't think it can be concluded that the evaluation is completed. I think it needs some further (management). I cannot evaluate how long this will take because (I am not around the model).

O. Hov: One might say that the sensitivity of the model tolerates realistic changes in model input parameters like deposition velocities, emission field, transport direction, wind speed, as it has been performed for this one episode, but the need clearly exists to perform more sensitivity studies, such as a study on the effect on the very simple way horizontal diffusion is included as well as vertical mixing. There should be additional sensitivity studies to acquire a feeling for the sensitivity of the model towards realistic changes in the model process.

D. Jost: We will take note of this.

O. Hov: As to plans for evaluation, it might be sufficient to consider some sensitivity studies that might be done this year or in 1984.

D. Jost: How about the other models?

P. Misra: Some evaluation will have been done for the ADOM model by 1986.

D. Jost: The SAI model?

S. Reynolds: We have conducted evaluations on portions of the model. I say "portions" because we have conducted these studies in the context of SO_2 and sulfate, but that uses transport. So, the physical process is part of the oxidant model. A study has been done and a report of it will be appearing in Atmospheric Environment. Also, we presented some results at this meeting for an 8-day application that was done in the Northeastern United States. This application is part of an evaluation going on at this time.

D. Jost: Thank you. Is it being done for SO_x?

G. Whitten: For oxidants as well. It was presented at this meeting.

D. Jost: Then I misunderstood.

G. Whitten: Yes, the oxidant results were presented here; the sulfate results are being published.

D. Jost: So the evaluation for your model will be done in 1984.

G. Carmichael: We will be doing some calculations of field studies in 1984, but the entire verification will be operational in 1986.

D. Jost: This would be understandable from those numbers. With respect to input data, I think the emissions data will be handled later on, so we will not look at them at this moment. We will limit ourselves to the meteorological data. Perhaps we can indicate for the European case what routine meteorological data will be sufficient for application of the model.

O. Hov: With respect to input data, I would like to suggest that OECD recommend that a unified approach be taken when the emissions inventories are established, so that the inventories from the various countries can be compared.

D. Jost: I wold like to say that the emissions inventories will be handled separately in addition to this table, and we should keep this in mind. Therefore, I will ask that we restrict our discussion at this time to the meteorological data.

D. Jost: For the Hov model, it is sufficient. This cannot be answered for the Lamb model, as this model may handle each data set that is provided.

P. Misra: My answer is yes, but I would like Anton to comment on that.

A. Eliassen: It requires some work, but it is sufficient.

D. Jost: Yes, it will require some work to apply European meteorological data to the Lamb model as well. That is, meteorological input data. As you have 11 layers in the SAI model, can these layers be taken from European meteorological balloon measurements?

G. Carmichael: Yes, from the upper air. You have the same problems with some of the other models. However, the more frequency you have, the better off you are.

R. van Aalst: Does this exclude the cloud data?

G. Carmichael: I am not sure about the cloud data. They are harder to obtain.

D. Jost: Do you have available routine WHO measurements from cloud data?

G. Carmichael: In principle, yes.

D. Jost: Then, this will not be different from the United States?

A. Christie: A great deal of preprocessing will be required to get the cloud data into the scale we are discussing here in order to be able to compute the input data from the available data, regardless of whether the data are from North America or Europe.

D. Jost: With respect to the output data, I would like to ask for time scale, how the average data will average and also with respect to the model scales that results from the models as they exist now. This will be given for the Hov model (as it is a trajectory model).

O. Hov: An appropriate scale for the output is not an hourly average but 6- or 8-h averages or more that that. The longer the time period we average, the better the agreement I would think. Also, I would say that as a Lagrangian model, it is most practical when a limited number of receptor points are of interest. If you want to compute an Eulerian field with a Lagrangian model, that becomes very expensive.

D. Jost: A limited number of receptors, whatever the limit is.

O. Hov: Much less than the number of grid points, which is 39 times 37.

D. Jost: With respect to the Lamb model, the Misra model, and the SAI model, I assume that it will be 1 h?

G. Whitten: We have a graphics output that is continuous.

D. Jost: I think we could take this as 1 h--continuous and 1 h.

A. Christie: I think what you're talking about, there is a minimum time resolution. (Inaudible comment) ...daily averages or weekly averages. In other words, an analysis of the effect you are going to get may be based on either a short-term effect or an integrated effect.

D. Jost: That is right, but I do not think that we can decide or recommend this right now. Obviously, you may come to a decision later on--I think 24 h would be sufficient--and obviously the quantification of different models will change. There will be models that will be more suitable for 1 h than other ones. There is, for example, 24 h where all these models might be equal, sufficient.

Unidentified Speaker: It might be worthwhile to at least quantify these input data a little more, at least according to low, medium, or high input requirements or according to the degree of sophistication. In other words, does the wind field model needed to drive it represent the state of the art? If you want to put in things like liquid water profiles and really do a good job on clouds, then I would call that a fairly sophisticated method. So, I think we ought to go at least one step further on the input data.

D. Jost: Are you saying that we should split up this question on the meteorological data?

Unidentified Speaker: Not just the meteorological data. It affects other things as well as the geophysical data, the degree of sophistication in your emissions files and so forth.

Basically, for every data set that you are inputting, there are various degrees of sophistication for which these models were either originally developed or various degrees of sophistication that would be consistent with the degree of approximation inherent within them. So at this point, I would not want to go beyond much more than putting low, medium, or high numbers on them in terms of the effort it would take to develop the input field. However, it might be worthwhile to have more than just what is there.

D. Jost: Low, medium, or high concerning all the input data, with the exception of the emissions inventories?

Unidentified Speaker: You might want to quantify it in terms of the man-years that would be required to actually compare the input, to adequately apply that model.

D. Jost: Starting from routinely available data, which effort is necessary to prepare the data in such a way that they are suitable for the model?

Unidentified Speaker: That is just one suggestion.

D. Jost: There seems to be agreement on it. To make this task easier, I should perhaps start with the Lamb model since I was a member of that panel discussion. In the discussion, I got the impression that the preparational work represents a high effort in terms of what we are discussing here. That is, you are starting from continuously available meteorological data, what you continuously know, topography, and so on.

R. Lamb: As far as the meteorological data are concerned, you can put in as much or as little data as you want You are limited by putting in one number or one observation for every grid cell, or you can put in a guess or assume a constant flow field. As for the way we do it in this simulation, the uncertainty and the error bounds on the calculation will depend on how much information you put in. So, you put in a lot or a little and you get out, in principle, reliable numbers or less reliable numbers.

D. Jost: This is why I was at first a bit afraid to put in more details as I saw. I guess that the complicated models are also simplified and then you have simplified. In the same sense, you may also simplify the input data and then it is no longer a simple or a complex model, but it is still possible.

H. van Dop: In constructing the complicated model, you have in mind a certain amount of meteorology. So, you need to say that you could put it as simple as you want it, but I do not think you really want that. You have in mind a certain sophistication of your chemistry too.

R. Lamb: That is true. On the one hand, having a lot of sophistication can allow you to handle the chemistry properly. You may need it for that. That is partly why this model has the resolution and the structure it does, to treat the chemistry, which is independent of the meteorology. So, for the flow fields that go into it, you would ideally like to have the maximum utility involved in putting as much meteorological information into it as you can get. For that reason, you want to have as much, so that would require a great amount of data, more than exists.

D. Jost: One could perhaps approach it from a (third). One could say that this model is meaningful for this model to use (a high effort) with meteorological data (where there could be models). There is no sense in making such a high effort, and then such a thing could be put here as high. It is possible and it is meaningful to spend much effect in preparing the input data for this model.

R. Lamb: I would agree.

O. Hov: I would like to comment on the meteorological input to the first model. As it is now, it operates on the same data base as is used in the normal weather forecast procedures. The trajectories are computed at this level in Oslo and uses the meteorological data along the trajectories that come from the weather forecast data base. So, I would say that the data base is fairly extensive, but that it is a very simple job to get those data to the model. So that's a low, work effort drops.

P. Misra: My first reaction would be a high effort, but again I would like to have Anton comment on how quickly he can get the data.

D. Jost: Do you object to this?

A. Eliassen: No.

D. Jost: What is you comment on this?

S. Reynolds: I think medium to high, depending on your definition. Less than some, more than others.

J. Killus: Since the Carmichael model has in fact many more layers and much more detail, the degree of input that you would expect is high-plus.

H. van Dop: Are we finished with input data?

D. Jost: I assume.

H. van Dop: I would like to go back to another point, which was discussed rather fast, I think, That is the output data. The 1-h time resolution I object to. Maybe it is true, but it is not thought out well.

D. Jost: Your point is right, but when I considered the 1 h, I thought that the people that are working with this model are meaningful to it, that they do not output 1-h averages knowing that there cannot be any difference between them.

H. van Dop: If you have a grid spacing of 150 km, it would be very useful to depict your data every hour. Every 3 h may be more meaningful.

D. Jost: This takes us to the discussion we had this morning on how to integrate reactions and all that happens within the grids. Perhaps we should keep this in mind, as I do not think that I will be able to assess this right now.

H. van Dop: Could we ask each modeler to indicate whether his model is supposed to give useful information every hour?

D. Jost: I assumed you would phrase the question a little differently. I do not think that any of the authors will say "No."

E. Runca: In describing the output, we should also consider the spatial scale of every model.

D. Jost: Yes, this could be added. The spatial scale of the output from the model is used right now.

As you mentioned during your discussion this morning, it is not too meaningful to discriminate hours--to distinguish scale hours and something like 100 km.

A. Eliassen: The emissions data are for 150 km.

D. Jost: I would like to put these numbers here in addition. The times--the scale for the output for your model?

R. Lamb: What is the number now?

D. Jost: Do you mean the local scale for the output of the model?

R. Lamb: The grid scale?

D. Jost: Yes.

R. Lamb: It is in latitude/longitude coordinates, but it is about 18 km.

D. Jost: 18 km.

R. Lamb: 18.5 km.

Unidentified Speaker: It is 50 km on the European scale.

D. Jost: Why is it different for the European and U.S. scales?

P. Builtjes: Small country.

D. Jost: I know that everything in this country is larger; that is why they are expressed in miles instead of kilometers. The SAI model?

J. Killus: The SAI model has been exercised both at the 20-km level and at the 80-km level.

D. Jost: 20 to 80 km.

G. Whitten: The SO_2/sulfate that we are doing is at 80 km and the seabreeze that I showed is designed at 8 km.

A. Eliassen: As to the spatial scales you just wrote down, noone can convince me that you can calculate oxidants for Europe or anywhere on a 20-km scale, because the wind fields and the other meteorological information are far too uncertain. Noone can convince me that you can say exactly with what probability you can do this.

Now, I would like to ask OECD what it really wants here. Do you want to calculate oxidants on a 20-km or a 50-km scale for Europe, or do you want to prevent the occurrence of high concentration of oxidants? Whether these high concentrations hit one city or a neighboring city, does that really matter? On an international scale, if the plume hits Belgium or The Netherlands, does that really matter?

In Europe, you have high emissions of different species coming out in the air, and these mix over a number of days. If you have a puff that is emitted from one source and it does not exactly hit the source 50 km down in that direction, it will hit another source 10 degrees off in another direction. Whatever you do, you more or less get oxidant production. So, it seems to me completely unnecessary to do this on such a fine scale, as some people here are proposing. You might also argue that this could result in a suggestion to reduce sources over certain regions or whatever and so on. This would have consequences for the weather next year. What sort of situations will you have then? The development of the weather is so uncertain that all sorts of cases can occur. This therefore limits the degree of accuracy that can be of interest at all when you apply models for oxidant control strategies.

Having listened to the discussion on all these very complex models, I have to say this. I have defended complex modeling at this meeting over the previous few days. Now, I resume my usual position of supporting the more simple models. Thank you.

P. Lieben: I think maybe the question here, I, not being an expert, feel that if the accuracy is 20 km, it can also be run for 50-55 km.

A. Eliassen: Of course. Any model is--

P. Lieben: The other way is not common, but provided you can get 5- or 10-km accuracy, you can also get 50- or 55-km accuracy. Is this right?

A. Eliassen: Yes, that is right.

P. Lieben: Again, just to comment on your point about these figures, it will be useful, once the OECD member countries have better defined what they would like to use as control strategies and, again, I say it is not for the group here to do it and to tell them what they should do that will help the countries to decide what to do. Once they have more ideas about the kind of control strategies they will try to achieve, this will probably help in looking at the final selection of a model. If they have to do it on a grid of 20 km x 20 km or higher but if they have to do it, then again that's information.

G. Whitten: As a chemist, I think that these models can be used to provide an accurate estimate of the effectiveness that an HC control strategy might have over an NO_x control strategy--an estimate of which would be more effective. It is true that we cannot predict the meteorology for future years, so we do not know exactly where or how big the O_3 will be. However, with a typical meteorological field, a typical emissions inventory, and a good model, we can predict the order of magnitude of the response from a control strategy in terms of HC versus NO_x and in terms of mobile sources versus stationary sources. This is what the models are being used for at the urban scale and they can also be extended to the mesoscale.

Unidentified Speaker: I just wanted to point out a few things that went into my comment about 50 km. Almost all of these models can be applied on any grid size you choose. As for what went into that 50 km, we looked at a map of Europe and decided we wanted to put the boundaries in fairly clean areas to the extent possible. So, that means that you run out the western boundary to include England, the northern one to include southern Scandinavian countries, etc. With the eastern boundary, you are in rough shape, but you can only do what you can do. So you pick an approximate scale for a model and consider how many grid points you can afford to run with. Now, 30 and 40 grid points in one direction (X and Y) are the kinds of numbers that people run with. If you say 50 or 60 in one direction, that is an extremely ambitious model; if you say 100, the model might not run on present day computers. So, even though they sound very arbitrary, that is the way in which these number get back down to the problem.

J. Bottenheim: If you are going to control HCs, there will be some consequences for the acid rain problem. I know you do not want to talk about acid rain, but the people who do research in acid rain are certainly interested in what you decide here for oxidant control, because it influences their decisions as well. So, you are right, Gary. However, there are consequences in other fields. I am not sure if you should only stick to your own little niche of oxidants.

S. Zwerver: I do not think this is a question of policies or abatement strategies. I cannot imagine that the same abatement strategies or policies will work for all countries. What is one country to do for abatement or the other. Here, it is just a question of the model itself; it has nothing to do with policy. I think there are two different points, strategies, but the question here has to do with the quality of the model. So if we stick to the issue of quality, whether the model needs a 1 x 1 square, we can accept that.

D. Jost: Is there any further discussion?

H. van Dop: The spatial resolution in the model is up to the modeler. The modeler has to decide which spatial resolution is needed for his/her numerical schemes, for his/her chemistry. Nobody cares if it is 1 m or a 100 km. The question is: What spatial scales do we want to have numbers for control strategies. Do we want every 10 km? Do we want to test every 10 km in our area or do we want 100-km values?

D. Jost: As Zwerver mentioned, there are several reasons for those scales. One reason is inherent in the model. The other involves local-scale concerns as effects. Or, due to a particular air quality situation, you may know that typical oxidant situations need a local resolution of 1 km, 10 km, or 50 km. By different aspects, we mean which scale you want to use to abate air pollution. This is, I think, quite different from this.

H. van Dop: It is in part a technical question because you have to ask the same question (as to the time required of the grid), at what distance do you expect a significantly different value from the source point.

D. Jost: I think this is given by this 10, 50, 80 km. I assume that the numbers that I mentioned with respect to the time scale are meaningful. Perhaps you normally should take half of the scale and look for differences in order to--It should not be the same scale, but half the scale. That is, there should be real differences when these numbers are doubled. But, are you proposing further items of a more administrative nature that should be taken?

A. Venkatram: Perhaps this is a very naive question, but what is a control strategy? What are the types of questions that comprise a control strategy? Do you have to pose the questions before you can discuss the utility of the models or answer those questions? What is a typical control strategy? Do you need to be specific about it so that you can match the model's answers to the questions you pose, especially with some of the models aspiring to predict probabilities. I think it is quite critical to ask these questions.

B. Luebkert: We are not trying to propose one typical control strategy, but we all asking whether you can use these models altogether to develop control strategies and we are evaluating different ones against each other to see what effects they have on the oxidant picture on a regional scale.

A. Venkatram: Do you have to ask the model a certain number of questions before it can give you an answer?

B. Luebkert: I thought we were trying to do that.

D. Jost: Didn't your question in this direction deal with control? For example, a typical strategy would be to decrease the NO_x emissions from high stacks and in the same strategy decrease the HC emissions from the motor traffic.

A. Venkatram: Qualitative.

D. Jost: Those could be enough to--

A. Venkatram: Is that a qualitative statement? People are looking for something like a 30% reduction in HCs to lead to a 30% reduction, for example. However, those percentages mean nothing unless you specify time scales, spatial scales, and probabilities.

A. Galli: That is true, but it is a lot easier for the U.S. to define those kinds of things than it is for the European countries. The U.S. has a standard that is based on 1-h time periods. We also have a standard of 0.12, which means that we are aiming for a percentage reduction to meet a particular number. The European countries do not have a corresponding level or regulatory basis with which to work. Therefore, it is a little harder for them to define that kind of information here when they have not defined it for themselves, for Europe or for a country within Europe. We more or less have defined that in the United States, at least for O_3, and our primary and secondary standards are identical, right now at 0.12.

A. Venkatram: If you work on the numbers, you can reduce the HCs by 20% and you might see an increase in O_3 next year because of the meteorology. This has to be posed in terms of probability. What is the probability of a decrease below a certain level?

D. Jost: In my opinion, such a result would help to say it is possible to do something for oxidant air pollution, and there are several possibilities to do this. Yes, I agree with you. Nevertheless, I think that it is necessary to base all of the very simple measures on more complicated scientific studies.

P. Lieben: What is your conclusion from the discussion regarding the best candidates for future focus by OECD?

D. Jost: I cannot give you a clear answer. To select one model or several models, we need some more basic information on the possibilities which we will have as it was discussed several times on the needs. To put it in a more qualitative way, I think it would be worthwhile to do some simple tests, first to run a test with one of the not-so-simple models in order to check several emission scenarios and then to check the model, or a third model that has been used, against a more complicated one, which would really take into account almost all the known chemical and physical effects. Although I cannot nominate one very simple model to do the whole business, I propose such a stepwise procedure, including testing, developing strategies, and checking the model sensitivity for areas in which the model is going to be used.

P. Lieben: Perhaps somebody in the audience would respond to the question. From the discussions we have had these 3 days, are there best candidates for the future by OECD?

J. Schjoldager: The comparison was very interesting, but from an applications point of view in Europe, a few things are missing. One important thing was brought up by Smith in terms of a home in Europe. These models really have a home in Europe. As far as I am concerned, three of them are now available and a fourth is underway. It is very important to consider which of these models has a home in Europe.

Unfortunately, the models you already have in Europe are quite different--the U.K. model, the Hov model, and the SAI model. Then, there is the Misra model, which will definitely have a home in Europe.

Given that and given Gary Whitten's comment, which is that we should not really restrict the discussion to one model, with which I fully agree, we should allow for a group of models to be used. I think these two things together early indicate that we should really not try at all the various models we have at home in Europe. We should more or less continue the discussion here along the lines that we should prepare the necessary data bases so that all of these models can be continued by the people involved with them. Thank you.

P. Lieben: Thank you for that very useful comment. It is time to go on to the third item, which may be the most difficult point, that is emissions inventories. I would like to ask Dr. Galli to introduce the subject for discussion.

A. Galli: Emissions inventories can perhaps be looked at in four different ways. The first one brings up a problem with what Joergen said. He talked about models finding a home. The first question that occurs with emissions inventories is: What compounds do you measure? You have several choices. The U.K. model and some of the other models require 40 specific compounds. If you are going to do an emissions inventory in Europe and if you are going to upgrade your inventory, you will have to look at all 40 compounds; otherwise, such a model will be useless to you. Furthermore, the more compounds you are looking at, the more expensive your emissions inventory will be, and the commitment on the part of the country will have to be a lot higher. While I understand the rationale, you immediately run into a problem gathering the data base for your emissions inventory, which illustrates the first question right here on your list: Exactly what types of compounds or materials are you going to measure? Are you going to measure for the 40 compounds that are identified in the brief compound, or are you going to limit it to a more reasonable number, which may be somewhere between 1 and 10?

I have indicated some likely candidates, and I am asking for suggestions and comments. I have suggested NO_x and HCs. In the case of HCs, what species are you talking about? I have also included O_3. The relationship between O_3 and acid rain has been discussed here, and it cannot be ignored. So, I have to include SO_x. What else? What about the other 36 compounds that are mentioned in some of the other models? Do you really want to talk about that from a European standpoint when, in essence, the place where your data bases are really going to need to be updated, and most of you have identified that as a problem in Europe. Do you want to do it for 40 compounds?

J. Bottenheim: Are you talking about just field measurements or source inventories?

A. Galli: I am generally talking about emissions inventories from sources, not ambient monitoring.

J. Bottenheim: If you are talking about inventories, it might not be as disastrous as you picture it.

A. Galli: I do not know; I am asking the question. I do not live in Europe; I live in North America.

J. Bottenheim: Neither do I, but apparently it has been done for the U.K. with some success. Maybe they can say something more about it.

K. Brice: Surely the question here about whether you speciate the HCs does not come down to what the model can handle. If you parameterize the chemistry, you still have to have as much information as possible about the HC composition. I think everybody would agree with that. For emissions, there are enough data available in some cases to be able to break down the HCs into more than just five or six categories. The more emissions you have, the better. In the field, measuring all of the HCs is of course more of a job. You are dealing with a lot of concentrations. It is a more extensive field project. So, I would argue that the more information you can get on the HCs, the better your parameterization of any chemical scheme.

A. Galli: I guess I was hearing from the group in general that the HC data base was the weakest of the European data base. If what you are saying is true, you really have a bigger problem than what you are describing, because what you are saying you need the most is your weakest area.

P. Builtjes: I said the HC field data are missing.

J. Killus: The more experience you have with emissions inventories, the more possible it is to make assumptions concerning the various speciation that occurs. Given an emissions inventory that is split into source-emission categories without precisely defining what HC properties are in those sources, it is still possible to make intelligent guesses as to what those new speciations are.

If you have mobile sources, for example, it is very difficult to go to a local gas station, run a few drops of gasoline through a gas chromatograph, and find out more or less what the unburned gasoline should look like. That generally applies across the board. If you then make a certain number of ambient speciation characterizations, you can compare the emissions inventory speciation to the ambient speciation.

For U.S. inventories in the early days, inventory speciation and ambient speciation did not correlate well at all. In more recent times, they have correlated quite well. In a recent urban study for Philadelphia, there was an extremely good match between the ambient speciation and the emissions inventory we were given. For that, we were profoundly grateful.

A. Galli: Are you saying that we should be looking at both an ambient inventory as well as an emissions inventory for specific sources?

J. Killus: Ambient measurement for quality assurance.

A. Galli: Any advice on the size of the list of the compounds?

R. van Aalst: If you are going to include acid rain components, I strongly suggest that you include ammonia. Our estimates for The Netherlands include a contribution of about 35% of ammonia compounds to acid deposition.

A. Galli: I am not sure whether acid rain compounds should be included. Personally, I have tried to make a clear distinction between the oxidant-type things and the acid rain, realizing that there is some relationship and that you cannot draw a distinct line between them. How about some suggestions of other compounds we have looked at?

B. Luekert: I would like to see the discussion separated into two different areas, first a discussion on emissions inventory and later one on ambient data. There is a lot of confusion about this, and I would like to make this distinction. If we first focus on the emissions data, I think the problem in Europe is that you have to agree on the methods to develop a common inventory. This is exactly what we are looking for--some suggestions, a basis that the countries come up with an inventory that fits whatever model will be selected or that fits different models.

However, it has to be compatible. There is no need for one country to use different emission factors than another country. When you do, you introduce mistakes into you model that your can predict right in the beginning. This is where we are looking for some recommendations.

S. Reynolds: You might want to consider CO.

G. Whitten: Carbon monoxide can also play a quality assurance role in validating a model. The chemisty of CO is slow; therefore, following CO clouds is a way of testing meteorology and testing against the emissions inventory.

G. Whitten: Our experience with emissions inventories to date suggest that a common problem is carbonyl compounds, aldehydes especially. Many of the chemical mechanisms respond to these types of compounds, so we are talking about oxygenates as opposed to pure HCs.

Our problem was that we asked for HCs. People were putting out oxygenated HCs and saying that these were not HCs. So, they did not list them. However, the oxegenated HCs still reacted in the atmosphere. That is why we are now using the terminology "VOCs and reactive organics." These have a particularly strong reactivity in the early morning when the sun comes up. If they are missing from the inventory, the model does not perform correctly.

L. Lindau: In response to Barbara Luebkert's comments about the emissions inventories in Europe, I agree that we have to develop them in a similar way for each country. However, the emission factors will be different. A refinery in one country is different from a refinery in another country. The emissions factors will be different and will have to be considered in some source-by-source way. The way of doing it, the principles, etc., have to be discussed.

H. van Dop: Should particulate matter be included?

R. van Aalst: Acid rain substances are not included; SO_2 should be removed as well. We agree on that.

J. Bottenheim: Just like O_3, yes?

A. Galli: What are you taking off?

J. Bottenheim: Take off O_3 and SO_x if you do not want to talk about acid rain, O_3 at any rate.

G. Whitten: We do not really know of any O_3 emissions.

B. Luebkert: To continue the discussion, we could consider some of the countries that have undertaken a joint, cooperative effort, such as the Dutch-German experience. The people here who have had experience with that may want to point out the problems they have run into with the inventories, the differences that exist between these countries. These experiences could be a learning process so that OECD would not repeat the same mistakes, but take it from there and avoid mistakes in the beginning.

D. Jost: There has not been much experience of this type up to now. We had a lot of time lag in preparing our emissions inventories just for the Dutch-German effort. One of the reasons was that we needed to know at a very early stage of the project which emissions data, what accuracy, what time and scale, and what resolution were needed. It took a lot of time to prepare this information. This had to be discussed with a lot of political people and already for this discussion one needs to know what the need of the single data one has. There is no possibility for change later on, as one has to follow the entire procedure all over again.

J. Killus: The SO_x should probably be placed back on the list. I can at least come up with a rationale to include it in photochemical episodes, for not looking at it specifically in terms of acid rain. After all, we tested the transport sections of our regional model using the SO_2 and sulfate portions of that, especially the SO_2/sulfate reaction that is a moderately good tracer for photochemial reactivity and for regional transport.

I am not really familiar with the various reasons for wanting to separate the two issues of acid deposition and regional oxidant modeling. However, for transport reasons and in order to test for transport calculations, I believe we should include acid rain.

A. Galli: You have to include acid rain only from the standpoint that it tends to be a buzz word within a number of countries for getting additional research support. The group here has more or less steered clear of acid rain, because it is being taken up in a number of other foreign and domestic forums. In these forums, the discussion is focusing directly on acid ran and only peripherally on oxidants and their effects upon acid rain.

It is not a hard and fast thing; it is something that helps to sell research programs within the individual countries. As for specifically talking about acid rain here, there are enough other forums that are bigger from both a technical and a political standpoint, forums where that is being taken up now, without adding this meeting as another one.

J. Killus: I am not talking about acid rain at all though. In fact, the regional oxidant episodes are not generally rainfall episodes. However, there are reasons for wanting to have SO_2 and sulfate in the transport model in order to get the test to do long-range transport. It is good validation situation and unless there is some reason to exclude it, I think it should be included.

P. Lieben: Maybe the question is a bit ridiculous, since the OECD countries are engaged anyway in making a detailed emissions survey of SO_x. So, it will be included anyway with this project or another projet. We will match the truth with the same grid square. However, there may be specifc reasons for including SO_x in an oxidant poject. That is where we need advice.

A. Galli: We are not trying to specifically exclude anything. I am saying that I do not want the acid rain, acid deposition, or whatever term you are using today, to be the driving force of this group, because there are many other areas where it is the driving force.

S. Reynolds: I might just comment on the experience that I perceived in the Dutch photochemical modeling effort and their assembling of an emissions inventory. They were able to develop a gridded inventory in a fairly expeditious fashion, given the data bases they had assembled there, including the derivation of the kinds of HC speciation that the photochemical model required. That was about a four-to-six lumped-species inventory. They had quite detailed estimates of the HC composition for various sources in their country.

A. Galli: I am not sure I know where you are heading.

R. van Aalst: It is probably a detail, but I might add that in The Netherlands at the moment we are concentrating a little bit on emissions in water as well. It may be a point that you forget when you are trying to get emission inventories; you may just concentrate on air emissions, although organic species, especially volatile organic species, tend to come from water. It is a detail that could be significant.

J. Bottenheim: I wanted to make another comment on other European situations. You say you are already developing an SO_x inventory; what other inventories are you developing? If you are already obtaining one for NO_x, why should you consider doing it in this group as well, and similarly for HCs or CO. In other words, it is nice to say that you want to stay with the oxidants, but if you are developing these inventories for the acid rain program, even if somebody else does it, it is rather superfluous to consider starting this effort again.

P. Lieben: No, the emissions survey for NO_x and HCs has not been fixed so far. We are trying to get some ideas about the grid square to be used and so on, from this particular meeting.

B. Luebkert: I would like to point to one application, especially the Dutch-German effort in that experience. You said that you had no problems in getting a gridded inventory. Was that only on a grid base or was that also with respect to source categories?

I think OECD is talking about eventually making some recommendations with respect to policy for control strategies. If so, then it is of no use to have only a gridded inventory that is needed by source categories. This is where there may be a source problem. If you come across any of those, I would like for them to be discussed.

P. Builtjes: It is available for all kinds of source categories. No problem at all.

S. Zwerver: I could not follow the discussion completely. As to the question of categories, it is, in principle, a very difficult problem to solve. In Europe and in OECD, we also have examples of miscategorized inventories for NO_x and HCs. For the Dutch situation and I suppose for the German situation, we were able to categorize in a very detailed way.

As I understand it, the problems now under discussion are the emissions inventories, the long-term average emissions inventories, and the oxidant models that will be applied, especially to short time periods (e.g., days).

Another problem arises: What are the emissions, the meteorology, etc., for that specific date? We have heard from the United States; they have very complicated inventories to collect data directed especially to the application of these short-time-period models.

I think that will become in Europe to gather information for certain periods, because that would mean a high degree of organization and so on, but I do not think that--

A. Galli: That is not my understanding of the discussions that have occurred over the past 3 or 4 days. My understanding is that it would be difficult to come up with these inventories. Maybe I am naive because I have not been there, but I believe there is an inventory problem in Europe. However, I do not believe that it is of such a level that these data could not be gathered in a 1-, 2-, or 3-yr time frame, the same time frame in which a lot of these models could be updated and tested in something like this.

S. Zwerver: It depends on what you mean by emissions inventories. For Europe, it is possible to relate, in a very simple way, energy use, energy consumption, etc., in order to make an emissions inventory, but would that inventory be sufficient? Do we need more detailed emissions inventories, directed to special periods.

P. Builtjes: In principle, the data are available. It is in time. But what you want to know is the traffic. So there are traffic grids specified by day. This information is available. You can even have specific episodes in mind. You can even try to look up specific situations. So you have to check, for example, where the large emitters--when they are working.

You can just by inquiring look to power plants to check whether specific things happenend, like an enormous strike in the petroleum industry or something, but that is not difficult. You have to organize it. It will take some time to do. You would really have to check whether there are strikes, you know. But that's not a difficult procedure.

J. Novak: One other area that was briefly mentioned up in these discussions was biogenic species. Right now, there is nothing on our list for biogenic HCs. In my opinion, you were talking about all anthropogenic sources. So, I would at least question whether this is important, whether it is something that Europeans would like to consider in their inventories and whether the chemistries will handle them exclusively or just lump them in with the other anthropogenic HCs.

P. Builtjes: As far as I know, the chemistry does not explicitly handle it. So the HCs coming out of these sources are categorized in the same way as the other emissions. But emissions factors, whether they are accurate or not (is not a question). But emissions are available, so you know by land use this 2-km^2 grid gives emission of so many--because it's grassland or because it's urbanized, etc.

J. Novak: Are you saying that biogenic emissions inventories could be constructed?

P. Builtjes: They are available. They are existing on the list, on the file. Because the land use is on the file.

J. Novak: In biomass factors?

P. Builtjes: Yes.

G. Whitten: What can be done explicitly in the chemistry is not always clear. If you handle them specifically in the emissions inventory and you bring them in and out of the emissions inventory, you get a differential effect in the results from your model of the biogenic emissions. Certain types of chemistry would need to be treated explicitly; other types could be treated in a lump. It would not make any difference.

A. Galli: Perhaps we should move on a bit. The next thing we need to discuss is the grid resolution. To a certain extent, the grid resolution we should be looking at was covered by Dieter in the previous discussion. In the various models, the resolution varied anywhere from--what did you say, eight-and-a-half, Greg?

G. Carmichael: Yes, that was for seabreeze.

A. Galli: I think it was 80 km in one case. We have heard one argument that the 20-km grid resolution is too small and rather meaningless. You need something a little bit larger, but I guess if you have a grid resolution at the smallest level (20 km), you can always do a resolution at 40, 50, or 60 km because the basic information is available. Do we really need to discuss grid resolution further?

S. Reynolds: It would perhaps be helpful if an inventory were put together in the future on a somewhat finer resolution than might seem appropriate at the moment, so that the inventory might be available at a subsequent time. I am thinking of something in the range of 24 km to 50 km. A hundred kilometers might be a little gross for some subsequent time.

G. Whitten: If you were going to use an urban model on a mesoscale, you would want a concentrated emissions inventory for that area.

P. Grennfelt: I would like to point out that there is one cell for Southern Europe. If this is too large, why not divide it so we get four squares? That would make it a little easier to compare the sulfur emissions. To construct a new grid system would be difficult in many countries to understand why you are using a new system.

P. Lieben: For example, your proposal would be 60 km to 65 km. So, if we put four squares together, we have one square of the existing grid.

P. Grennfelt: Yes.

A. Eliassen: It seems to me that the main reason for going to a very fine grid is related to chemistry.

If you want to go just a short distance downwind from an industrial area, say 12 h, you might say that it is important to have very good spatial resolution for the emissions in that area. The further downwind you go, perhaps the less important the emissions resolution is. So, it is difficult to say how important this is, and it is difficult to discuss it here without having good data to base your arguments on.

I suggest that you perform calculations with a very coarse emissions grid, say 150 km, then a very fine grid, and that you qualify them by your calculations, with a realistic presentation of the different species in the fine grid. Then, compare the results. That would be a very interesting guiding experiment and it would perhaps tell OECD what resolution is really necessary in this case.

P. Lieben: That idea is interesting, but it would just postpone any decision about the grid size to use.

A. Eliassen: Just because there is no basis on which to decide that now?

P. Lieben: Well, I was thinking that we could have some ideas, some proposals from the meeting, I am sorry to come back to this question, but I think it is a very important point.

I have heard during these 3 days that in any model development project, especially, the emissions data are costing the most. Once we have the emissions data, once we have put forth the effort to get it, we will not do it again for another grid size resolution for a long period of time. So, I think it is important to get an idea of which size we should propose.

J. Bottenheim: I am not sure if the size is really relevant if you start making an inventory. If you make an inventory, you try to figure out where the point sources are and you establish coordinates for them. You go out to where the area sources are and you establish coordinates for them. Then, it is up to the user to divide them and add them up in a certain grid size, and I think the major effort goes into determining where these sources are on the map and putting coordinates on them. Whether you next add them up on a 20-km grid or on a 100-km grid is up to the user, and it is presumably a relatively simple effort compared to just coming up with where those sources are, with the coordinates. In other words, I think this grid discussion is rather irrelevant.

D. Jost: I agree. If you are establishing an emissions inventory and if you are going beyond a scale of several kilometers, say 5 or 10 km, there is not much difference in the cost between using the 10-km grid scale or the 100-km grid scale, since you will have to rely on the same data.

On the other hand, it is true that we do not have much data here before us. However, the data we have seen are more or less of a typical scale for the O_3 concentration, somewhere in the range of 20 to 50 km. The whole plume has a scale of 90 km or something like this.

B. Luebkert: I am not sure that it is very easy to go from a large grid to a smaller grid. It is easy to go from a smaller grid to a larger one, but if you agree on something like a 100-km^2 resolution and later find that you need a 20-km resolution, a major effort will be required to take into account area sources. Certainly you have point sources, coordinates on the map, but area sources are calculated from land use patterns, etc. So, you would need to do that exercise over again. For an area like Europe, that is quite costly.

J. Bottenheim: You are basically suggesting that area sources over 100 km not be done. In my opinion, if you do that, you do not take too large an area anyway. You still need--for areas sources, you would divide it up on smaller units anyway. If you do not want to use it, then you don't have to add them up.

A. Galli: I do not think at this point that we (or OECD) have really made any decision based on which way to go. OECD is gathering opinions, information on which to make recommendations to the member countries, who may or may not approve the suggestions given to them that are based on the conversations coming out of this workshop. So, your comments are well taken and there are other people here who have supported these comments, but they have to be considered in the context of the total discussion we are having here. They may be accepted or they may be rejected down the line, depending upon the opinions of the different countries, but I do not think a decision is being made here at all.

F. Smith: I think we do have to decide upon a scale. We do have to recognize Anton's point when we are dealing with nonlinear chemistry, that no absolute scale can be chosen that solves the problem.

A. Galli: Right.

F. Smith: When we are dealing with the interplay of different chemical species, the only scale that really matters is almost a molecular scale. Of course, there is no way we can get down to that. So, at some stage we have to parameterize what is going on at the molecular level. The former parameterization may in fact be a function of the actual scale that we choose. Recognizing that, the most logical scale to impact here is perhaps related to the size and the variation of the sources.

A. Galli: Any other questions? Let's move on to the next problem--the domain, in other words the boundaries for your region. As I have perceived from prior discussions, this has been more of a problem to the European community and than it has been to North America, including the U.S. and Canada.

I have heard two things mentioned in term of the European community: (1) the boundary problems between Eastern and Wester bloc nations and the inability, to date, to really obtain emissions and source information from the Eastern bloc and (2) the identification of an emissions inventory for a specific area, whether that area is some subset of a country or a country plus a little bit of another one.

We need to hear comments on how the European area might be handled or how an emissions inventory might be handled for the European-type situation. Also, we need to hear suggestions for problems on the North American continent.

P. Lieben: This has to be looked at both in connection with the models we have considered and in connection with the models that may finally be retained for use, since a model might perform better than the others for a limited part of Europe than it does for the whole system.

D. Jost: As mentioned several times, we have the Dutch-German cooperative effort in which modeling of this oxidant problem has already been studied. As we expect to measure relatively high O_3 concentrations in the southern part of our country, it would be necessary for us to include in this OECD project larger parts of our country, larger parts in this Dutch-German effort. Obviously, the Dutch-German effort and the experience we acquired could be used as a basis for determining what needs to be done within the OECD project.

P. Lieben: Any comments on that?

A. Galli: Let's go on to the last point, meteorology. You are talking about two types of observations, surface observations right at the land and observations of the upper atmosphere. It has been suggested that we include wind speed, wind direction, temperature, humidity, etc., the things that you routinely look at. In the upper atmosphere, you are looking at inversion layers, anything that most modelers would not ordinarily consider as part of their meteorological information.

L. Kropp: I have a question regarding an issue that has already been discussed but I missed the answer. Did we agree to a certain time resolution for the emissions inventories. Should it be 1 h, 1 day, 1 yr?

A. Galli: I do not think that we made any real decision.

L. Kropp: I think it is an issue that must be resolved before establishing--

A. Galli: I agree, and once this meeting is over, OECD will be putting together some recommendations based on the comments and information that have been presented here. The only comment that I made on the subject was that it is little bit easier for the U.S. to define the time period, since it has a regulation that is essentially based on 1 h. The European countries really do not have regulations based on a particular time frame. So, with the U.S. it is 1 h. It is defined; it is known. With the foreign countries, it is not that way. Many of them do not have a time period.

J. Bottenheim: I disagree with you. This is precisely the answer that should come out of this workshop. Some of the models will need source inventories on an hourly basis if they are ever going to produce hourly answers.

A. Galli: I am not saying it should not come out.

P. Builtjes: For traffic, it is on an hourly basis.

J. Bottenheim: If you want to run a model and get hourly answers to validate your model, you need hourly inventories.

P. Builtjes: Sure.

J. Bottonheim: That is a major problem that has been found in all of these models. They try to predict 1-hour, 6-h, or 1-day values and use yearly source inventories. What does that mean? It means that most of the uncertainty comes from the source inventories. If OECD is just going to recommend source inventories on a yearly basis, there will be certain consequences for whatever model you are going to use or validate with. This is a very important point that has to be discussed here.

A. Galli: I do not have a problem with that, and I do not mind it being discussed here. I did not say it was unimportant. I said that this forum is not making the final decision.

P. Lieben: That is a very important point, and I agree. It must be looked at in relation to the model to be used. The emissions inventory has to match the model requirements. If they do not match, you can do nothing.

A. Galli: I could not agree with you more; that has to be decided.

D. Jost: I agree that we need such emissions inventories, but there is some misunderstanding here. None of the known emissions inventories is in a real-time 1-h resolution that processes timed hourly values. That is not too hard, but getting real-time 1-h emissions inventories is impossible and not really necessary.

Also, this very fine emissions inventory from the Northeast Corridor produces hourly values, but they are not actual hourly values. They are processed based on some assumptions on the emissions. This would be possible for Europe, too.

G. Whitten: I am not quite sure I followed the discussion. If you are talking about meteorology, you have a 100-km grid size. The meteorology with that mass of air cannot do a lot in 1 h; it cannot move very far. So, there is less of a constraint on the time scale of the meteorology from that standpoint. You have this huge air mass and there is no way that it is going to move very far in 1 h. However, the sun can go up and come down a lot in 1 h. There are certain parts of the model that you do have resolution for.

A. Christie: It seems to me that, by using 15-min time steps, we are talking about insulation changes that may have a fairly massive emission (of every hour). You average that over a 1-h or a 3-h period and you say it has not moved very far, but it may have a significant effect in any Eulerian model. While you may not need that kind of resolution when it comes to using the model for strategy evaluation, you will probably need it to carry out an evaluation on the model, at least for short time periods. We have already discussed the evaluation of these models, and it seems that we cannot carry out an adequate evaluation without that kind of resolution over at least a limited period.

J. Novak: I was just going to make a couple of comments again about the emissions resolution. I think it is a good idea for OECD, as part of its recommendations, to list those specific parameters that it might want to include in relation to the emissions inventory. As Peter was saying, most inventories in the U.S. are collected on an annual basis, on a county-wide basis for area sources.

The types of statistics are exactly that, about different fuel uses or whatever it might be. I think it would be worthwhile to go through that exercise in Europe, looking at the kinds of statistics that are available to do temporal distributions and sending somebody out to look for that type of information.

Also, it would be worthwhile to try to characterize the differences between the countries. It was mentioned before in terms of the different emissions factors, the different refineries, and other things in terms of the fuel types that are used. The differences that could occur need to be specified on a country basis. I think you will frequently find that these are uniform and that you can isolate those that are not uniform and handle them in specific ways. So, an important part of actually producing your emissions is looking for this additional information and finding out exactly what is available.

P. Lieben: Thank you. Are there any other comments on the emissions inventory input data?

G. Whitten: I might make one final comment from the chemist's standpoint and cite some experiments that were done at the North Carolina Outdoor Smog Chamber on behalf of the Australian Government. They were quite concerned with a spectrum of solvents that seemed to have different HC composition. So, several smog chamber experiments were conducted with a wide range of solvent compositions. The result was that the reactivity of the solvents was all about equal, and they all produced about the same rate and the same amount of O_3 for a given weight of HC.

The point is that, even though there might be a different brand of gasoline in one country than in another, a different car spectrum, or things like that, you see changes amongst a large amount of HCs. The result to date is that, if you have a wide spectrum of HCs, once you have one spectrum, they all seem to react very similarly. We have not yet seen too many important differences. So, this kind of thing becomes unimportant, the fine structure and the reactivity spectrum.

P. Lieben: Thank you. Are there any other comments on this point? It seems we have more or less exhausted the possibilities.

Well, I would like to thank you very much for all these comments. I would like to turn the meeting over to our general chairman.

D. Jost: As we are nearing the end of this meeting, I would like to thank Basil Dimitriades and our colleagues from the United States, especially the U.S. Environmental Protection Agency, for inviting us, for making possible this meeting, and for providing the scientific background to help us come a little bit closer to the definition of our project within OECD. Again, thank you very much.

B. Dimitriades: You are quite welcome, and thank you very much.

I would just like to say a few things. First, when the decision was made in November to hold this conference in April, I thought it would be a hopeless undertaking and that the time we had to prepare was not nearly enough to tackle this truly complex subject, but I am delighted that the discussions went so well. They were well thought out and informative. When all of this information is digested, I feel that there will be some very concrete and useful conclusions to be turned over to the OECD.

I would like to thank the speakers and the experts, who were invited to come here and help us on such a short notice. They graciously accepted, and they did an outstanding job. A lot of credit also goes to the other participants. Their critical comments in the discussions made the conference as successful as it turned out to be.

Speaking for myself and the other members of the EPA staff from ERC and from Washington, we are delighted for this experience.

I want to say once again that we seriously invite one or two modelers from OECD to spend a year or so with us in our laboratory, working with us in the regional modeling area. Please give it some thought, and we can talk about this again.

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